Optical pumping of the electron spin polarization in bulk CuCl

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In CuCl bulk crystal negatively charged excitons (trions $X^-$) can be induced by the resonant optical excitation of extra electrons in conduction band minimum. In the case of circularly polarized light and due to the top valence band structure only the electrons with one spin orientation (for example antiparallel to the direction of the light propagation in the case of $\sigma^+$ polarization) contribute to the formation of trions $X^-$. At the same time spontaneous decay of $X^-$ populates both, parallel and antiparallel electron spin states, thus producing an enhancement of one spin orientation. We propose to use this mechanism for optical pumping of electronic spin polarization. We estimate the momentum matrix elements of electron - trion transitions which are the main factors determining the pumping rate. The pumping dynamics is described by the density matrix evolution equations which couple to the Maxwell wave equation for the coherent pumping laser pulse propagating through the sample. The results of our simple model calculations suggest that spin polarization close to 100% can be achieved in CuCl in the time scale of the order of 100 ps.

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I. INTRODUCTION

In many semiconductors the spin lifetime of an electron at the bottom of the conduction band is much longer than the electron effective mass. The electron spin, as a carrier of information, has big potential in many possible spintronics and quantum informatics applications. A basic requirement for the workable devices is the possibility of sufficient initial electron spin polarization. One of the known methods is the injection of the electrons into semiconductor media through the spin aligner (magnetic or semimagnetic contact) [1, 7]. Recent achievements of this technique claim 85% efficiency [4, 5]. Another way is the optical excitation of semiconductor media with circularly polarized light which creates spin polarized electrons and holes [8–11]. For example, in bulk GaAs, the representative zinc-blende direct gap semiconductor, the ratio of densities of excited electrons with opposite spin polarizations reaches 3:1 due to the optical selection rules for the transitions from heavy and light hole subbands. In low-dimensional structures, because of the split-off of the heavy and light hole subbands, the degree of electron spin polarization can in principle reach 100%. However, the spin polarization of electrons excited from the valence bands is transient. The time of its persistence is limited mainly by the lifetime of electrons which can recombine with holes. The electrons and holes separation by an electric field is needed in order to extend the spin lifetime.

In the case of single electrons in quantum dots (QDs) many possible ways of spin initialization and manipulation have been proposed. Among them several interesting proposals refer to optical control [8, 9, 10–12, 13, 14]. The electron in QD can be resonantly excited into the heavy hole trion $X^-$ state [12]. Shabayev et al. [10] suggest to use the trion as an intermediate state for feasible initialization of the electronic spin. In order to obtain the well defined spin polarization, regardless of its initial state, they propose to use circularly polarized optical $\pi$ pulse combined with $\pi$ pulse of transverse magnetic field and successive spontaneous trion decay.

One can think about using the same or similar mechanism as proposed for QDs for electronic spin polarization in the case of small concentration of excess free electrons in semiconducting quantum well structure or bulk material. However, the binding energy of trion $E_{b2}$ (i.e. binding of the second electron to the exciton) in bulk is usually much smaller than 1meV and even in 2d quantum wells it rarely exceeds 1meV [13, 16–18, 19, 20]. making the selective creation of trions rather difficult. Furthermore the top of the valence band is fourfold degenerate in the typical zinc-blende semiconductors and due to the selection rules for the trion creation it would be rather hard (but not impossible providing the trion’s spin relaxation time is long enough in comparison to the trion lifetime) to reach high electronic spin polarization in the bulk material. Nevertheless in the special case of copper halides, because of the very small ratio of electron to hole effective masses $\sigma = m_e/m_h$ in these materials, the binding energy of $X^-$ can appear to be relatively large. In CuCl, which is widely used for direct creation of biexcitons and recently also for generation of entangled photons [21], the effective mass of the hole is high ($m_h \approx 20m_e$), and because of that $E_{b2}$ is expected to be 6 meV [22, 23]. CuCl also has not typical structure of the top of the valence band (the top subband is twofold degenerate). This in principle makes possible to obtain electronic spin polarization close to 100% by optical pumping and without using additional magnetic field pulses. To reach this goal we propose to illuminate the sample with circularly polarized light of appropriate frequency, tuned to the electron trion transition. During
the illumination electrons with only one defined spin projection on the direction of the light propagation can participate in the trions creation, while the electrons having opposite spin projection are not affected by light. When the coherent light is used, the populations of trions and participating electrons oscillate with Rabi frequency. Because created trions can spontaneously decay to electrons with both spin projections, the densities of electrons and trions participating in oscillations diminish after some time in favor of rising density of electrons non affected by light. This, in turn, enhances the spin polarization.

In order to estimate the trion lifetime which influences the rate of the spin pumping, we have used the effective mass approximation to describe the trion bound state and for the calculation of appropriate matrix elements for optical transitions. The methods used, the results of the calculations and relevant selection rules are given in section II. In the section III we explore the problem of the pumping dynamics in the presence of the light pulse propagating through the sample using the density matrix approach and Maxwell equation. We consider charged sample with small amount of excess free electrons, which are not generated by band to band transitions. Such situation may, for instance, correspond to extra carriers introduced by injection or by contact with n-doped semiconductor and/or simultaneously applying electric field. We assume that the temperature is close to zero and we deal with the fully occupied valence band before and after pumping.

II. FREE ELECTRON ↔ TRION X− TRANSITIONS

CuCl has direct band gap $E_G = 3.43$ eV at $k=0$ and tetrahedral symmetry $T_d$. The lowest conduction band of symmetry $\Gamma_6$ and the uppermost valence band of symmetry $\Gamma_7$ are both twofold degenerate at $k=0$. A lower fourfold degenerate valence band of symmetry $\Gamma_8$ is split off by the spin orbit interaction by $\Delta(\Gamma_7 - \Gamma_8) = 69$ meV. The $\Gamma_6$ and $\Gamma_7$ Bloch states correspond to the angular momentum $J = 1/2$. Expressing the electron wave functions with the symmetry of $s$, $p_x$, $p_y$ and $p_z$ orbitals as $S$, $X$, $Y$ and $Z$ respectively, the conduction Bloch functions $\Gamma_6$ can be written as

$$c_{1/2} = S|\alpha\rangle, \quad c_{-1/2} = S|\beta\rangle \quad (1)$$

and the valence Bloch functions are of the form

$$v_{1/2} = \frac{1}{\sqrt{3}}[(X + iY)|\beta\rangle + Z|\alpha\rangle],$$

$$v_{-1/2} = \frac{1}{\sqrt{3}}[(X - iY)|\alpha\rangle - Z|\beta\rangle], \quad (2)$$

where the indexes $+1/2$ and $-1/2$ denote projections $m_j$ of the total angular momentum of band electron onto $z$ axis, while $|\alpha\rangle$ and $|\beta\rangle$ denote the parallel and antiparallel to $z$ projections of pure electron spin.

The trion $X^-$ is formed by the three quasi particles, two electrons and one hole, interacting with the Coulomb field. In absence of an applied magnetic field the negatively charged exciton has only one bound state corresponding to the two electron spin singlet $|2\rangle$. Then we assume the bound state of $X^-$ to be a linear combination

$$|X^-; \mathbf{K}, M_j = \pm 1/2\rangle = \frac{1}{\sqrt{2}} \sum_{k_1,k_2,k_h} C(k_1,k_2,k_h) \times$$

$$(a^+_{k_1+1/2}a^+_{k_2-1/2} - a^+_{k_1-1/2}a^+_{k_2+1/2})d^z_{k_h,\pm 1/2}|g\rangle, \quad (3)$$

where $\mathbf{K}$ is the total momentum of $X^-$, $|g\rangle$ denotes the electronic state corresponding to the empty conduction and fully occupied valence band, and $a^z_{k,m_j}(d^z_{k,m_j})$ denote the creation operator of an electron (hole) in the Bloch states with the wave vector $k$ and projection of the angular momentum $m_j$. In the effective mass approximation the linear coefficients $C(k_1,k_2,k_h)$, subjected to $k_1 + k_2 + k_h = \mathbf{K}$, are the Fourier transforms of the trion $X^-$ envelope $\Phi(r_1, r_2, r_h) \sim \exp(iKR_0)\psi(r_{1h}, r_{2h})$, where $R_0$ denotes the center of mass vector of the whole complex $X^-$, while $r_{1h}$, $r_{2h}$ are the relative coordinates of the two electrons (1, 2) and one hole ($h$).

In order to investigate the free electron - trion transitions we calculate the appropriate dipole matrix elements in a similar way as given by Stebe et al [23]. For the electric dipole transition between the trion state with $M_j = +1/2$ and the free electron states $a^z_{k,m_j}|g\rangle$ with $m_j = \pm 1/2$ we get

$$\langle g|a^z_{k,m_j}\exp(-iqr)|X^-; \mathbf{K}, +1/2\rangle =$$

$$\frac{1}{\sqrt{3}} \langle Z|\mathbf{p}|S\rangle I(\mathbf{k}, \mathbf{q}) \delta_{\mathbf{K}, \mathbf{k}+\mathbf{q}} \quad (4)$$

for $m_j=+1/2$, and

$$\frac{1}{\sqrt{3}} \langle X - iY|\mathbf{p}|S\rangle I(\mathbf{k}, \mathbf{q}) \delta_{\mathbf{K}, \mathbf{k}+\mathbf{q}} \quad (5)$$

for $m_j = -1/2$. The factor $I(\mathbf{k}, \mathbf{q})$ depends on the electron and emitted photon wave vectors $\mathbf{k}$ and $\mathbf{q}$, respectively, and is defined as the integral

$$I(\mathbf{k}, \mathbf{q}) = \sum_{l} C(\mathbf{k}, \mathbf{q} + 1, 1) =$$

$$\int_{V} \exp[-i\mathbf{kr} + (1 - \mu)|\mathbf{q}|\psi(\mathbf{r}, 0)d^3r, \quad (6)$$

with $\mu = (1 + \sigma)/(1 + 2\sigma)$ (the formula (4) coincides with that given by Stebe et al [23] when $\mathbf{q} = 0$).
Due to the selection rules of the dipole transitions resulting from the Eqs. (4) and (5), there are two channels of spontaneous decay of $X^-$: first, corresponding to the transition $X^- \rightarrow e_\uparrow$ with the rate $w_1$, and the second one, corresponding to the transition $X^- \rightarrow e_\downarrow$ with the rate $w_2$ (hereafter for the trion $X^-$, as well as for the electron $e$ we use the arrows $\downarrow$ and $\uparrow$ to denote the angular momentum projections $-1/2$ and $+1/2$ onto $z$ axis). Obviously, the same rates correspond to $X^+_\downarrow$ - decay, i.e. $w_1$ for $X^+_\downarrow \rightarrow e_\downarrow$ and $w_2$ for $X^+_\downarrow \rightarrow e_\uparrow$. Due to $T_d$ symmetry of CuCl, the matrix elements $\langle X | \hat{p}_x | S \rangle$, $\langle Y | \hat{p}_y | S \rangle$, $\langle Z | \hat{p}_z | S \rangle$ are all equal to each other and from Eqs. (4) and (5) it follows that the ratio of rates is $w_1/w_2 = 1/2$.

In order to estimate the values of the rates $w_{1,2}$, we have calculated the integral $I$ with the envelope $\psi(r_1h, r_2h)$ approximated by the calibrated wave function of the $H^-$ ion, where the wave function of $H^-$ was taken as proposed by Rotenberg and Stein [24]. Such an approximation can be justified by the very small value of $\sigma = 0.02$ in CuCl. The obtained value of $I^z$ (called the envelope oscillator strength) decreases much slower with $k$, than $I^2$ obtained by Stebe et al [27] for $\sigma = 0.1$ or $\sigma = 1$. At $k = 0$ we have obtained $I^z = 676$, which is very close to the value obtained for small $\sigma$ in ref. [24]. Using the experimental data: $|\langle X | \hat{p}_x | S \rangle|^2/m \approx 3$ eV, $\hbar \omega \approx 3.2$ eV (energy of emitted photon), $\eta \approx 1.9$ (the refractive index) and integrating over all photon directions we estimate that the rate

$$w_1 \approx \frac{4e^2 \omega \eta}{9 \hbar c^2 m^2} |\langle X | \hat{p}_x | S \rangle|^2 \approx 1.2 \cdot 10^{11} \text{s}^{-1} \ , \quad (7)$$

and the radiative lifetime of the trion $\tau = (w_1 + w_2)^{-1} \approx 2.8 \text{ ps}$.

In the case of resonant transitions induced by coherent illumination, the electron and trion densities oscillate with Rabi frequency

$$\Omega = 2dE/\hbar \ , \quad (8)$$

where $E$ is the electric field modulus and $d$ denotes the magnitude of the dipole moment of the electron - trion transition $d = \sqrt{2/3} |\langle X | \hat{p}_x | S \rangle| |e|/m \omega$.

III. SPIN PUMPING

Let us consider as a pumping light the $\sigma^+$ circularly polarized coherent plane wave pulse with frequency $\omega$ and electrical vector $\mathbf{E}(z,t) = \sqrt{2}E(z,t)\hat{x}\cos(\omega t - qz) + \hat{y}\sin(\omega t - qz)$, entering and propagating in a sample along $z$ direction. The shape of the envelope $E$ can change in time due to the coherent coupling of the light with electrons and trions. For the $\sigma^+$ polarized light transmitted along the $z$ axis the only possible induced transitions are the transitions between electron states with $m_j = -1/2$ and the trion states with $M_j = +1/2$.

FIG. 1: (a) Induced (wavy arrows) and spontaneous (solid arrow) transitions between conduction electron and trion $X^-$. (b) The possible electron states (dots) to which trions can recombine spontaneously, defined by the momentum $(\mathbf{K} = \mathbf{k'} + \mathbf{q'})$ and energy $(E_{X^-}(\mathbf{K}) = \hbar \omega' + E_e(\mathbf{k'})$) conservation.

The $\sigma^+$ signal does not affect the electrons with $m_j = +1/2$, because the trion $X^-$ with $M_j = 3/2$ does not exist. Trion $X^-$ has a short lifetime and can recombine spontaneously into the free electron states. As it is described in the previous section the trion $X^-$ with $M_j = +1/2$ decays spontaneously into the electron state with either $m_j = +1/2$ or $m_j = -1/2$, with the relevant rates $w_1 = 1/3\tau$ and $w_2 = 2/3\tau$, respectively. So under the influence of $\sigma^+$ polarized light the density of electrons with $m_j = +1/2$ increases, while density of electrons with $m_j = -1/2$ decreases, with analogy to the standard optical pumping of electron spins in atoms, which is based on the depletion of one of the ground-state sublevels and accumulation in the other spin sublevel [22]. The $w_1/w_2$ ratio is given by the symmetry properties of the wave functions. We should emphasize however that the pumping process described here can succeed for any $w_1/w_2$ providing it is not too small. If bigger is $w_1$ the faster is the pumping rate.

During the decay of trions the photon transition energies fulfill the equation $\hbar \omega = E_{X^-}(\mathbf{K}) - E_e(\mathbf{k})$, where $E_e(\mathbf{k}) = h^2 k^2/2m_e$ is the energy of electron in conduction band, $E_{X^-}(\mathbf{K}) = E_{X^-} + \hbar K^2/2(2m_e + m_h)$ is the energy of the trion with $\mathbf{K} = \mathbf{k} + \mathbf{q}$ and $E_{X^-} = E_G - E_{b(\text{ex})} - E_{b2}$ ($E_{b(\text{ex})}$ denotes binding energy of a free exciton). The photon energies lie below the threshold $\hbar \omega \approx E_{X^-}$ (see Fig.1a) and correspond to the photon wavelength $\lambda \approx 384 \text{nm}$. If the Fermi momentum $k_F$ is sufficiently small in comparison to the photon wave number $q$ (i.e the Fermi energy of the excess electrons is respectively smaller than $h^2 q^2/2m_e = (\hbar \omega)^2 \eta^2/2m_e c^2 \approx 0.09 \text{ meV}$, what corresponds to the concentration of electrons $N < N_0 = 10^{15} \text{ cm}^{-3}$) then practically all electron states to which trions can recombine, are available (see Fig. 1b). Moreover, if the spectral width of the pumping light signal $\Delta \hbar \omega$ is close to the trion linewidth ($\approx 0.22 \text{ meV}$), corresponding to its short lifetime, than all photons in the pumping pulse can participate in the pumping process. Since the
oscillator strength $I^2$ does not change significantly with $k$ up to a few values of $q$, it follows that almost all electron states, to which trions recombine spontaneously, can be involved again in the process of spin pumping with similar probability. It can happen that after multiple processes of induced transitions and spontaneous decays some electrons can be moved in the Brillouin zone far from its center and reach the places where they do not participate any longer in the process while still being in the state $m_j = -1/2$. However, we can safely assume that the probability of such an event to occur in the time scale of few tens of ps, which is typical time scale for electron energy relaxation processes in semiconductors, is very small. In the consequence, we can assume that all electrons with spin down can participate in the spin pumping.

Besides the induced coherent Rabi oscillations and incoherent trion decays also the incoherent interaction of electrons and trions with the environment should be taken into account and we describe them in our model with the phenomenological parameters. In our model we do not take into account the mobility of the free electrons and trions, and what follows, the diffusion of electronic spin, since it can be neglected in the timescales given by speed of light as well as by trionic spin lifetimes. We assume the long spin lifetime of an electron at (or near) the bottom of the conduction band, however we take into account the possible spin flips of trions. The total spin of trion is determined by the spin of hole. We expect that because of the strong spin-orbit interaction for electrons in the valence band the hole spin lifetime $\tau_h$ in CuCl is short and of the order of few picoseconds (We do not know the exact hole spin relaxation time in CuCl, but we assume that it is of the order of typical time in cubic semiconductors). We expect the same for the spin lifetime of the trion, and assume that it is equal to $\tau_h$. Taking into account the possible trion spin flips, we consider the system of four (two electron and two trion) states. The scheme of all the possible transitions between these states is shown in Fig. 2. We would like to note here, that the trion spin flips do not interfere with electron spin pumping but, on the contrary, they open the second competing channel for the pumping process. What’s more, because the state $|X^-\rangle$ decays spontaneously into the electron state $|e_\uparrow\rangle$ twice as fast as into the state $|e_\downarrow\rangle$, the spin flips of the trions do not delay the electron spin pumping.

We describe the dynamics of the electron spin polarization with the density matrix operator $\dot{\rho}$ depending on time and position in the sample and written in the basis of two electron and two trion states. The time evolution equation for $\dot{\rho}$ in the rotated basis $|e_\uparrow\rangle, |e_\downarrow\rangle, |X^-\rangle, |X^+\rangle$, with respect to the laser light frequency $\omega$, is given below (the diagonal elements of $\dot{\rho}$ are labelled with single index):

$$
\dot{\rho}_{e\uparrow} = \frac{1}{3\tau} \rho_{X^-\uparrow} + \frac{2}{3\tau} \rho_{X^+\uparrow} \\
\dot{\rho}_{e\downarrow} = -\Omega Im\rho_{e\downarrow,X^-} + \rho_{X^-\downarrow} \cdot \frac{2}{3\tau} + \rho_{X^+\downarrow} \cdot \frac{1}{3\tau} \\
\dot{\rho}_{X^-\uparrow} = \Omega Im\rho_{e\downarrow,X^-} - \rho_{X^-\uparrow} \cdot \frac{1}{\tau} - (\rho_{X^-\downarrow} - \rho_{X^+\downarrow}) \cdot \delta \\
\dot{\rho}_{X^-\downarrow} = -\rho_{X^-\uparrow} \cdot \frac{1}{\tau} + (\rho_{X^-\downarrow} - \rho_{X^+\downarrow}) \cdot \delta \\
\dot{\rho}_{e\downarrow,X^-} = \frac{i}{2} \Omega (\rho_{X^-\uparrow} - \rho_{X^-\downarrow} - \rho_{e\downarrow,X^-} \cdot \gamma) ,
$$

where $\gamma$ is the transverse dumping rate and $\delta = 1/\tau_h$. The time evolution Eq. (9) is coupled to the Maxwell equation for the electric field by the relation

$$
\frac{\partial E}{\partial z} + \frac{1}{v} \frac{\partial E}{\partial t} = \frac{2\pi q N d}{\eta^2} Im\rho_{e\downarrow,X^-} ,
$$

where envelope $E$ is a slowly varying function of $z$ and $t$ and the right hand side of Eq. (10) corresponds to the extra polarization due to the induced transitions between the free electron and trion states. The refractive index $\eta$ (and the velocity of light in the sample $v = c/\eta$) is assumed to be unaffected by the light intensity, as the plasma frequency for considered free electron concentrations $\omega_p = \sqrt{4\pi Ne^2/m}$ is lower than $10^{-4}\omega$.

We consider a CuCl sample with the small volume electric charge. Let us imagine for example, that the surface plane $XZ$ of the sample is in contact with appropriately chosen n-doped semiconductor. It provides electrons that can be additionally driven into CuCl crystal by the applied voltage in the $y$ direction. We assume then, that in the thin layer, parallel to the $XZ$ plane, at the certain fixed distance from the contact, the electron charge distribution can be treated as constant. We relate considered above spin pumping process to such a layer. In the practical realization of the proposed experiment it would be important to take into account the real charge density distribution dependent on the way the electrons are injected and the geometry of the sample. Also the effects of the electromagnetic wave scattering at sample boundaries and interfaces should be taken into account.

In our first attempt to describe the process we neglect those effects and restrict our study to the general properties of the model.
We assume, that the initial density of trions is equal to 0, initial electronic spin densities \( (n_\uparrow) \) and \( (n_\downarrow) \) are both equal to \( N/2 \) and electrons are in the mixed state \( (\rho_{ee} = \rho_{e\delta} = 1/2) \). In Fig.3 and 4 we present the solutions of the Eqs. (8), (9) and (10) for two types of illumination and various input data. Fig.3 presents the influence of the passing Gaussian light pulse on the electronic and trionic spin populations (the appropriate diagonal density matrix elements) for the chosen time values measured since the moment when maximum value \( E_0 \) of the envelope \( E \) enters into the sample. It can be observed that during the illumination the Rabi oscillations between \( e_\uparrow \) and \( X_\uparrow \) are induced with the frequency \( \Omega \) dependent on the temporary value of \( E \), as given in Eq. (8). The energy stored in the pulse is continuously dissipated due to the spontaneous recombination of trions and finally the light pulse dies at certain depth. The energy dissipation causes also the lowering of the speed at which the pulse travels through the sample. The results shown in Fig.3 present the case when the appropriately short light pulse, after passing the sample, leaves behind the nonmonotonic spin polarization. This effect is related to the nonmonotonic dependence on \( z \) of the final local Rabi oscillation phase.

Fig.4 shows four examples of the influence of the step-like shaped signal with \( E_0 \) amplitude on the electron and trion spin distributions (the step-like shape has been in fact kindly smoothed out within the narrow width \( v\tau \)). The signal enters the sample at \( t = 0 \). After few trion lifetimes the front of the travelling signal establishes its shape and all cases shown in Fig.4 relate to time \( t = 12\tau \) when the front shape is already constant. The same is true for the shapes of the spin populations. In the case of small dumping rates, Rabi oscillations can be observed just before the signal’s front. The light pumps electronic spins and makes the sample transparent. After examination of many results, obtained with various parameters, we have made some general observations. First, the speed of the signal front depends on the intensity \( E_0 \) and on the electron concentration \( N \), and can be much smaller than the speed of light in the crystal \( v \). Below some critical value of \( E_0^2/N \), the front speed decreases with rising \( N \) and with reduction of the energy (\( \sim E_0^2 \)) stored in the pulse (compare the position of the front in the Fig. 4c with its positions in Fig. 4a and b). Other observations are related to the speed of spin pumping. Since the moment when the signal front reaches the given position \( z \), the pumping speed is determined mainly by the trion lifetime \( \tau \), provided that frequency of Rabi oscillations \( \Omega > \tau^{-1} \). The speed of pumping is also not noticeably influenced by damping rates \( \delta \) and \( \gamma \), if their values are not higher than \( 10\tau^{-1} \). Nevertheless they influence the damping rate of Rabi oscillations (compare Fig.4c with Fig.4d).

The presented results of the simulations of spin pumping dynamics are obtained for \( \Omega \tau \leq 16 \), corresponding
to the density of photons in the incident laser beam $E^2/2\pi \hbar \omega < 10^{13} \text{cm}^{-3}$, i.e. to the laser intensity radiation less than 150kW/cm$^2$, and situates the intensity within the range used in the optical solid state experiments.

In our model we have not accounted for the depolarization of electron spins caused by the photons spontaneously reradiated by trions. This process can interfere with spin pumping and in general is hard to estimate. However, the density of the photons emitted spontaneously by trions and interacting with electrons is limited by the size of the space where electrons are located. For example, if the electrons are confined in the 2dim layer of the thickness $r$ and parallel to the XZ plane, then density of photons is lower than $NP_{X^-}r/v\tau$, where $NP_{X^-}/\tau$ is of the order of the density of the trions which decay in a unit of time. The rate of reabsorption of the photons in the process of recurrent creation of $X^-$ with electrons having spin ↑ is proportional to the density of photons as well as to the transverse lifetime $\gamma^{-1}$, which does not exceed $\tau$. Taken the above into account one can find that the depolarization of electronic spins is negligible under the condition $Nr < 5 \cdot 10^9 \text{cm}^{-2}$. Thus, when the thickness of the sample or the thickness of the space where electrons are located is $r \approx 1 \mu m$ the concentration of the excess electrons should be $N < 5 \cdot 10^{13} \text{cm}^{-3}$. The thickness $r$ should not be too small, in order to exclude the quantum confinement effects.

We have also neglected the effect of spin depolarization caused by the electron-trion collisions assuming, that the electron-trion scattering rates, similarly as the electron-electron scattering rates in semiconductor, are usually negligible in the case of small electron concentration, considered in our model. The elastic collisions between electrons do not change the electron spin densities and therefore they do not influence the pumping process. The same can be said about the elastic collisions between trions.

The separate remark should be made concerning the role of possible creation of excitons by the pumping light pulse. Even if the energy separation between trion and exciton is large, the high density of valence electrons in comparison with the low concentration of excess electrons can make the probability of excitonic creation comparable to the probability of the creation of trions. However, when excitons collide with electrons in inelastic way, then the second channel opens for trions creation. The examination of the appropriate selection rules makes clear, that exciton created by $\sigma^+$ optical pulse can form trion only with the electron in the state $|e_x\rangle$, which does not mean, that the new channel does not interfere with the first channel discussed here, and it can influence only the pumping rate.

Our model predicts that once the light reaches the given place then close to 100% polarization of electronic spins associated with full transparency of the crystal can be obtained there in the time shorter than about $20\tau$. Assuming that the sample has a size in the $z$ direction of the order of few mm and taking into account the time needed for the front of the light pulse to cross the sample, we can estimate, that in the case of high enough illumination all electronic spins in the sample can be fully polarized in the time scale of the order of 100ps.

In conclusion we propose the efficient and fast method of the electron spin pumping induced by coherent laser
light in the CuCl crystal. We have used several approximations for the estimation of the trion lifetime and for the description of the pumping dynamics. We have obtained the timescale of the process and the results showing interesting effects of pumping (like possible spacial modulation of spin polarization and slow down of the speed of the front of the light pulse). The incoherent interactions with environment have been described by using phenomenological parameters, which are not well known. We have examined the pumping dynamics in the broad range of these parameters observing that they influence the pumping rate very little, having however high impact on the dumping of Rabi oscillation. The spin pumping rate depends mainly on the $w_1$ rate (equal to 1/3 of the inverted trion lifetime). The proposed model can be used in the case of very small density of free excess electrons, which can be challenging to control in the experiment. In the possible experimental realization many factors which can influence the pumping dynamics, like real sample geometry, light scattering and interference effects as well as inhomogeneous electron concentration, should be taken into account. The degree of spin polarization can be evaluated in the experiment by measuring for example the transparency of the sample in the case of the illumination with circularly polarized light. CuCl has inverted (if compared to other typical semiconductors) valence band structure and relatively large binding energy of the negatively charged excitons. We expect, that similar electron spin polarization through the intermediate trion states is also possible, but probably not as effective, in other materials with typical valence band structure and usually having also much lower binding energy of trions. The efficiency of the process in these materials should be higher in two dimensional structures, where energy separation between trions and excitons is larger.

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