Optical control of electron spin coherence in CdTe/(Cd,Mg)Te quantum wells

1. INTRODUCTION

In this paper we report on a different regime, where the control effect is determined by additive contributions of the pump and control pulses. It is shown that in this regime the signal suppression is generated by resonant excitation of the negatively charged exciton (trion) resonance. We found that the control effect is enhanced by proper choice of the control pulse polarization. The developed quantitative theory allows us to explain these experimental data quantitatively.

The paper is organized as follows. After introducing the optical excitation with such a pulse does not lead to any spin polarization, we describe theoretical considerations based on a two-level model for the optical excitation of the resident electron to the trion.

Sec. V. Here we also discuss possible reasons for deviations between the experiment and theory at high control powers. We demonstrate that the polarization vector undergoes a full revolution on the Bloch sphere.

The experimental results are compared with the modeling in Sec. IV. We found that the control effect is determined by additive and non-additive mechanisms, whose relative strengths depend on the relative energy of the control and pump photons.

We demonstrate that the polarization vector undergoes a full revolution on the Bloch sphere. We conclude that the control effect is determined by additive and non-additive mechanisms, whose relative strengths depend on the relative energy of the control and pump photons.
II. EXPERIMENTAL TECHNIQUES

The studied CdTe/Cd$_{0.78}$Mg$_{0.22}$Te QW heterostructure (sample 031901D) was grown by molecular-beam epitaxy (MBE) on top of a 2-µm CdTe buffer layer deposited on a (100)-oriented GaAs substrate. It contains 5 periods, each of them consisting of a 110-nm-thick Cd$_{0.78}$Mg$_{0.22}$Te barrier and a 20-nm-thick CdTe QW. An additional 110-nm-thick barrier was grown on top of this layer sequence to reduce the influence of surface charges on the confined electronic states in the QWs. The barriers include 15 nm layers doped by Iodine donors, which are separated by 20 nm spacers from the QWs. These modulation doped layers provide electrons being collected in the QWs, where two-dimensional electron gases (2DEGs) with a low density of about $n_e = 2 \times 10^{10}$ cm$^{-2}$ form. This sample has a slightly larger electron density compared to its partner sample 031901C ($n_e = 1.1 \times 10^{10}$ cm$^{-2}$) grown on the same substrate by a wettable growth technique [12] which has been studied in Ref. [10].

The optical properties of both samples are, however, similar to each other, see Refs. [10, 13, 14] for details. The measurements were performed in magnetic fields up to 7 T applied perpendicular to the structure growth axis, $B \perp \mathbf{z}$ (Voigt geometry). The sample was immersed in pumped liquid Helium at a temperature of $T = 1.9$ K.

Time-resolved pump-probe Kerr rotation (KR) technique was used to study the coherent spin dynamics of the resident QW electrons [10]. Two mode-locked Ti:Sapphire lasers synchronized with each other generated the 1.5 ps pump and control pulses (spectral width of about 1 meV) at a repetition frequency of 75.6 MHz. The probe beam was split off from the pump laser, as sketched in Fig. 1(a). For the experiments reported here both lasers were tuned to the same photon energy corresponding to the trion resonance.

The electron spin coherence was excited by the pump and control pulses, for which different polarization configurations were used: The control was either co- or cross-circularly polarized with respect to the pump of fixed circular $\sigma^+$ polarization, or it was linearly polarized. The induced spin coherences were monitored by the reflected linearly polarized probe pulse, for which the angle of Kerr rotation was measured by a balanced photodetector interfaced by a lock-in amplifier, after sending it through a polarization sensitive Glan-Thompson beam splitter. The time delay between pump and probe pulses could be varied up to 7 ns by a mechanical delay line. A second delay line was used to set a fixed delay of the control pulse relative to the pump pulse. This delay could be changed up to $t_{pc} \leq 2$ ns in order to tune the phases of the spin coherences initiated pump and control with respect to each other.

Two protocols of pump and control beam modulation were used. First we present experiments, where the signals are mainly given by the additive effect of the pump and control actions. Here both pump beam and control beam were modulated by a chopper at a frequency of 1 kHz, so that the detected Kerr rotation signal reflects the effect of both beams. These measurements are described in Secs. III A-III C.

In order to study the “non-additive” effect of the control on the pump induced signal we used a protocol in which only pump beam was modulated. It was sent through a photoelastic modulator operated at 50 kHz frequency so that the polarization was modulated between $\sigma^+$ and $\sigma^-$. The polarization of the control beam was constant in time. The Kerr rotation signal was detected at the pump modulation frequency of 50 kHz, which allows us to suppress the additive contribution to the electron spin polarization induced by the non-modulated control beam. These results are reported in Sec. III D.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Photoluminescence (PL) and reflectivity spectra of the studied QW structure are shown in Fig. 1(b). The heavy-hole exciton (X) and negatively charged trion (T) resonances are clearly seen as minima in the reflectivity spectrum and as lines in the PL spectrum. They are separated by 2 meV, which corresponds to the trion binding energy [11, 15]. The broadening of these lines is mainly due to exciton and trion localization on QW width fluctuations. From the relative oscillator strengths of the exciton and trion resonances in the reflectivity spectrum we evaluate the resident electron concentration in the QW as $n_e = 2 \times 10^{10}$ cm$^{-2}$ using the method described in Ref. [10].

![FIG. 1: (Color online) (a) Scheme of the three-pulse time-resolved Kerr rotation experiment. (b) Photoluminescence and reflectivity spectra of a 20-nm-thick CdTe/Cd$_{0.78}$Mg$_{0.22}$Te QW. PL was measured under nonresonant cw excitation with photon energy of 2.33 eV.](image-url)

A typical Kerr rotation signal measured at a magnetic field of 0.5 T is shown in Fig. 2 by curve (a). The $\sigma^+$ circularly polarized pump pulse hits the sample at zero time delay and induces coherent spin precession of the resident electrons about the external magnetic field. The precession is reflected by the periodically oscillating Kerr signal amplitude $K(t)$. The oscillation period corresponds to the electron Larmor frequency $\omega_e = \mu_B g_s B / \hbar$ with an
electron g-factor \(|g_e| = 1.64\), which is in good agreement with literature data \([17]\). Here \(\mu_B\) is the Bohr magneton. The g-factor value was obtained from fitting the experimental data by an exponentially decaying harmonic function \([4]\)

\[
K(t) = A \exp\left(-\frac{t}{T_2^*}\right) \cos(\omega_c t).
\]

Here \(A\) corresponds to the signal amplitude, \(T_2^*\) is the dephasing time describing the signal decay. The evaluated dephasing time \(T_2^* = 4.2\) ns is considerably longer than the trion recombination times in the range of 30-100 ps in CdTe-based QWs \([10]\), which allows us to ascribe the Kerr signal to resident electrons.

A. Effect of circularly polarized control on signal amplitude

We turn now to the main topic of the present paper, namely the effect of control pulses, delayed by a time \(t_{pc}\) relative to the pump pulse, on the electron spin coherence generated by the pump. The modifications induced by the control depend critically on the reduced phase \(\varphi\) with which the control hits the pump excited electron spin coherence. This reduced phase is defined as \(\omega_c t_{pc} = \varphi + 2\pi N\), where \(N\) is an integer corresponding to the number of full spin precession periods during the pump-control delay, and \(0 \leq \varphi < 2\pi\).

We describe first the pump-probe experiments in which a circularly polarized control was used. We also focus on the signal amplitude modifications induced by the control, the changes of the phase are discussed in Sec. \(\text{III B}\). To that end we adjust the delay \(t_{pc}\) such that phase \(\varphi = 0\) is achieved, when the Kerr signal amplitude \(K(t)\) is maximum. At a magnetic field of 0.5 T this condition is fulfilled e.g. at \(t_{pc} = 0.87\) ns or \(t_{pc} = 0.96\) ns. The latter example can see in Fig. 2 by comparing curves (a) and (b). For co-polarized pump and control pulses (both \(\sigma^+\)) of the same power the Kerr signal is enhanced about twice after control action, see curve (d). This is the expected result, as in this case the electron spin polarization generated by the control has the same orientation as the one generated by the pump after a few full revolutions about the field. In contrast, cross-polarization of the pump (\(\sigma^+\)) and control (\(\sigma^-\)) pulses leads to full suppression of the electron spin precession signal, as shown by curve (c). In this case the electron polarizations generated by the pump and control are antiparallel and compensate each other.

Note, that for the low excitation density regime presented in Fig. 2 only a small fraction of the resident electrons is affected by the pump and control pulses. In this case the joint action of the pump and control can be described such that each of them generates spin coherence for two independent subensembles of electrons. The experimentally measured Kerr rotation signal results from their independent contributions, which make either additive or subtractive effect on the observed signal. Note, that a very similar behavior has been previously reported for the Mn spin coherence in CdTe/(Cd,Mn)Te QWs \([9]\).

Detailed results for the effect of control power on the Kerr signal amplitude for co- and cross-polarizations of pump and control are given in Fig. \(\text{III}\). The phase for control pulse arrival was chosen to be \(\varphi = 0\), as in Fig. 2. Therefore, the spin polarizations induced by the pump (\(S_{\text{pump}}\)) and the control (\(S_{\text{control}}\)) are either parallel or antiparallel to each other for co- and cross-polarizations, respectively. The resultant polarization (\(S_{\text{total}}\)) along the \(z\)-axis is reduced or increased, as shown schematically in the corresponding panels of Fig. \(\text{III}\).

The Kerr amplitude increases for the co-polarized configuration shown in Fig. \(\text{III}\) (a), in line with the intuitive expectations. It decreases for the cross-polarized case given in Fig. \(\text{III}\) (b), crosses the zero level when the control power becomes about equal to the pump power and then shows increasing negative values. These dependencies can be seen in detail in Fig. 3 where the dependence of the Kerr amplitude on control power is plotted. To determine the spin beat amplitudes the signals after the control pulse arrival were fitted by Eq. \(1\). Triangles and circles give the experimental data for co- and cross-polarized pump and control pulses, respectively. The absolute changes of the KR amplitudes relative to the dashed line are larger for the cross-polarized configu-
B. Effect of circularly polarized control on signal phase

When the control pulse acts on the pump induced polarization at an arbitrary phase \( \varphi \), not only the amplitude of the Kerr rotation signal changes, but also the phase will be shifted by an angle \( \theta \) after the control pulse arrival. Corresponding experimental data are shown in Fig. 3(a), where we chose cross-polarization for pump and control and \( \varphi = \pi/2 \). The inset in Fig. 3(b) shows schematically that for these experimental conditions the signal after the control pulse is expected to show a negative phase shift, i.e. to shift to earlier delays. The signal after control pulse arrival can be described by Eq. 1 when replacing \( \cos(\omega_t t) \) by \( \cos(\omega_t t + \theta) \):

\[
K(t) = A \exp \left( -\frac{t}{T^2} \right) \cos(\omega_t t + \theta). \tag{2}
\]

In agreement with our qualitative expectations, the signal phase shown in Fig. 3(b) by the filled circles decreases and saturates at \( \theta = -\pi/2 \) for control powers strongly exceeding the pump power.

The open circles in Fig. 3(b) show the signal phase evaluated from the experimental signal amplitudes without and with control using the simple additive model depicted in the inset of Fig. 3(a). As one can see from scheme the phase shift \( \theta \) is determined in this case of perpendicular orientation of \( S_{\text{pump}} \) and \( S_{\text{control}} \) by

\[
\theta = \arctan(S_{\text{control}}/S_{\text{pump}}). \tag{3}
\]

The overall tendency of the dependences shown by the closed and open circles is the same. However, they deviate considerably from each other for control powers exceeding 0.5 W/cm\(^2\). This evidences some non-additive contribution of the control to the spin coherence generated by the pump which we will discuss in detail below.

The results in Fig. 4 have been collected to confirm the conclusion drawn from the data in Fig. 3 that the phase shift of the Kerr rotation signal is mainly controlled by the ratio of the pump and control generated spin polarizations, \( S_{\text{control}}/S_{\text{pump}} \). An increase of the control power for constant pump power causes a shift of the signal to earlier times, compare curves 1 and 2. This corresponds to an increase of the phase shift value, as shown by the left diagram. In turn, a pump power increase for constant control power (curves 2 and 3 and the right diagram) induces a signal shift to later times. For the chosen power densities these transformations are dominated by the additive mechanism.

C. Effect of linearly polarized control

In our experimental geometry it is not expected that linearly polarized light would induce any spin polarization of the resident electrons. Indeed, we did not find any signal for a linearly polarized pump. However, we...
observed that the electron spin polarization induced by a circularly polarized pump is strongly sensitive to a linearly polarized control. One can see in Fig. 7 that irrespective of the delay $t_{pc}$ the Kerr rotation signal is suppressed by a linearly polarized control. The suppression effect increases for higher control powers as shown in the insert. One should note that this effect changes only the signal amplitude but does not induce any phase shift $\theta$, independently of $t_{pc}$. The suppression is clearly a non-additive effect: generation of spin coherence by the control pulse is absent, but the signal is still modified. These, at first glance, surprising experimental findings can be explained by the qualitative model presented in the following section.

D. Qualitative model consideration of linearly polarized control action

In order to develop a qualitative picture of the spin depolarization by the linearly polarized control we consider the simple model of a spin ensemble described in Ref. [10]. We represent the linearly polarized pulse as a superposition of two circularly polarized ones and assume that at the hit time of the control pulse there are $n_+$ electrons with spin $z$-component $1/2$ and $n_-$ electrons with spin $z$-component $-1/2$. We assume that the control pulse arrives at the maximum ($\varphi = 0$) or the minimum ($\varphi = \pi$) of the pump-induced spin beats, i.e. there are no in-plane spin components at the moment of control pulse arrival.

The absorption of the $\sigma^+$ component of the linearly polarized light generates $n_+ W$ singlet trions by exciting the same number of $s_z = +1/2$ resident electrons. Here $W$ is the probability of singlet trion formation per electron due
The electron ensemble after control pulse arrival is given by \( z \) respectively. The \( z \) component after and before the control pulse arrival, \( \leq \).

Clearly, the probability of singlet trion formation is 0.

One can see in Fig. 8 that also a circularly polarized control decreases the Kerr rotation amplitude, similar to the case of a linearly polarized control. The magnitude of this effect is identical for \( \sigma^+ \) and \( \sigma^- \) polarization of the control and is also independent of the control delay \( t_{pc} \) (not shown). It is interesting that the suppression efficiency of the circularly polarized control is equal to the one for a linearly polarized control of the same intensity. This suggests that the responsible mechanism is the same, which is confirmed by the quantitative analysis given below.

In Figure 9 the effect of the non-additive contribution is presented for various pump and control powers. The Kerr rotation signals are normalized to their maximum amplitudes before control pulse arrival. Two conclusions follow from these experimental data. First, the suppression efficiency increases with increase of the control power.

E. Non-additive contribution of control

In this section we address experimentally the question whether a circularly polarized control, similar to a linearly polarized one, can serve as a depolarizer of the induced spin coherence. This will also allow us to obtain in-depth insight into the non-additive contribution noted in Sec. III B. Our goal here is to study modifications of the pump-induced spin coherence by the control. For that one should exclude Kerr rotation signal that is directly caused by generation of electron spin polarization by the circularly polarized control. It is possible to suppress this signal by implementing the second measurement protocol described in Sec. II. Only the pump beam is modulated in this case and lock-in detection allows us to exclude the direct contribution of the unmodulated control to the detected spin polarization.

FIG. 7: (Color online) Kerr rotation signals measured at different time moments of control pulse arrival (indicated by arrows) for \( \sigma^+ \) polarized pump with \( P_p = 2.2 \) W/cm\(^2\) and linearly polarized control with \( P_c = 2.2 \) W/cm\(^2\). \( B = 0.5 \) T. Arrival times of the control pulses are shown by arrows: (1) \( t_{pc} = 0.82 \) ns, \( \varphi = \pi \); (2) \( t_{pc} = 0.95 \) ns, \( \varphi = 1.8\pi \); and (3) \( t_{pc} = 0.96 \) ns, \( \varphi = 0 \). Insert illustrates suppression of KR signal amplitude with increasing control power. The amplitude is normalized to its value without control.

FIG. 8: (Color online) Non-additive effect on Kerr rotation signals measured for different control polarizations (\( \sigma^+ \), \( P_c = 3.5 \) W/cm\(^2\) when only the pump beam is modulated (\( P_p = 0.25 \) W/cm\(^2\)). Arrow indicates the time moment of control pulse arrival at \( t_{pc} = 0.87 \) ns, \( \varphi = 0 \). \( B = 0.5 \) T.

FIG. 9: (Color online) Kerr rotation signal (arb. units) vs. time (ns) for various control delays and pump and control powers. The control signal is measured for \( \varphi = \pi \), \( \sigma^- \), and \( \sigma^+ \). The pump only signal is also shown for comparison.
power. Second, the suppression efficiency is determined by the control power only, compare the signal amplitudes for different pump powers before and after control arrival for the same control power of 3.5 W/cm².

![Kerr rotation signal](image)

**FIG. 9:** (Color online) Non-additive effect on Kerr rotation signals measured at various pump and control (σ⁺) powers (pump beam modulation only). Signals are normalized by the pump power to simplify comparison with each other. Thick solid line is pump only signal for \( P_p = 0.25 \) W/cm², \( B = 0.5 \) T, \( t_{pc} = 0.87 \) ns, and \( \varphi = 0 \).

### IV. QUANTITATIVE THEORY

The quantitative theory of spin manipulation by a control pulse is developed following the methods described in Ref. [18]. The electric field of the control pulse can be written as

\[
E(r, t) = E_{σ^+}(r, t)σ_+ + E_{σ^-}(r, t)σ_- + \text{c.c.},
\]

where \( σ_± \) are the circularly polarized unit vectors related to the unit vectors \( σ_x \parallel x \) and \( σ_y \parallel y \) by \( σ_± = (σ_x ± iσ_y)/\sqrt{2} \). Here the components \( E_{σ^+} \) and \( E_{σ^-} \) are proportional to the product of the exponential function \( \exp(-iω_c t) \) with \( ω_c \) being the control pulse optical frequency and a smooth envelope.

The incident electromagnetic field induces optical transitions between the electron state and the trion state, creating a coherent superposition of them. In accordance with the selection rules \( σ^+ \) circularly polarized light creates a superposition of the +1/2 electron and +3/2 trion states, while \( σ^- \) polarized light creates a superposition of the −1/2 electron and −3/2 trion states. In order to describe these superpositions it is convenient to introduce a four component wavefunction

\[
Ψ = (ψ_{1/2}, ψ_{−1/2}, ψ_{3/2}, ψ_{−3/2}),
\]

where the ±1/2 subscripts denote the electron spin projection and ±3/2 refer to the spin projection of the hole in the trion. The electron spin polarization is expressed in terms of \( ψ_{±1/2} \) as follows

\[
\begin{align*}
S_z &= (|ψ_{1/2}|^2 − |ψ_{−1/2}|^2)/2, \\
S_x &= ℜ(ψ_{1/2}ψ^*_{−1/2}), \\
S_y &= −3(ψ_{1/2}ψ^*_{−1/2}).
\end{align*}
\]

Here \( ℜ \) and \( ℑ \) are real and imaginary parts, respectively. All excited states of the system, such as e.g. triplet trion states are neglected. In this respect the model is directly applicable to the case of a resident carrier strongly localized in a quantum dot or quantum well imperfection. The role of excited states will be discussed below, in Sec. IV C.

Further, we assume that the delay between the pump and control pulses exceeds by far the radiative lifetime of the trion, hence, just before the control pulse arrival there is a resident electron with precessing spin but no trion. The state of the system just before the control pulse arrival corresponds to the non-zero components \( |ψ_{±1/2}|^2 + |ψ_{−1/2}|^2 = 1 \) and \( ψ_{±3/2} = 0 \).

Following the method in Ref. [18] and introducing smooth envelopes for the \( σ^+ \) and \( σ^- \) polarized components of the control pulse by

\[
f_{±}(t) = −\frac{e^{iω_c t}}{\hbar} \int d(r)E_{σ±}(r, t) d^3r,
\]

where \( d(r) \) is the effective transition dipole, see Eq. (12) in Ref. [18], one may reduce the Schroedinger equation for the four-component wave function to two independent differential equations for \( ψ_{±1/2}(t) \) which take the following simple form

\[
\dot{ψ}_{±1/2} = −(iω_0 + \frac{f_{±}(t)}{f(t)}) ψ_{±1/2} + f^2_{±}(t)ψ_{±1/2} = 0.
\]

Here \( ω' = ω_0 \) is the detuning between the control pulse optical frequency and the trion resonance frequency, \( ω_0 \). This simple form of Eqs. (9) follows from (i) disregarding other excited states of the system and (ii) neglecting the control pulse duration compared to the trion lifetime and the electron spin precession period in magnetic field. Below we discuss the cases of linearly and circularly polarized control pulses.

#### A. Linearly polarized control

In case of a control pulse linearly polarized along the \( x \) axis the circular components of the pulse envelope function can be written as

\[
f_{±}(t) = \frac{μ}{√2 \cosh(\frac{πt}{τ_p})},
\]

where...
where the factor $1/\sqrt{2}$ is introduced for convenience, $\mu$ characterizes the amplitude of the control pulse and $\tau_p$ is its duration. The pulse area is defined as $\Theta = 2\mu\tau_p$. The solution of Eqs. (9) can be recast as

$$\psi_{1/2}(+\infty) = \psi_{1/2}(-\infty)Q_i e^{i\phi_i},$$
$$\psi_{-1/2}(+\infty) = \psi_{-1/2}(-\infty)Q_i e^{i\phi_i},$$

(11)

where the constants $Q_i$ and $\Phi_i$ describe the transformation of the wavefunction under action of the linearly polarized pulse. For the case of a Rosen&Zener pulse, Eq. (10), one has

$$Q_i^2 = 1 - \frac{\sin^2(\Theta_i/2)}{\cosh^2(\pi y)},$$

(12)

where $\Theta_i = 2\mu\tau_p/\sqrt{2}$ is the effective area of each circularly polarized component of the control pulse, and $y = \omega'\tau_p/(2\pi)$. The expression for the constant $\Phi_i$ is rather bulky and is therefore not given here, see Eq. (26) in Ref. [18].

Using the definitions of the spin components, Eqs. (3), one can readily obtain from Eq. (11) that the spin vector of an electron after the control pulse, $S^{(a)}$, is connected with the electron spin vector before the control pulse arrival, $S^{(b)}$, by

$$S^{(a)} = Q_i^2 S^{(b)},$$

(13)

i.e. the spin vector before the control pulse is simply multiplied by some nonnegative quantity $Q_i^2 \leq 1$. If the electron is left behind unpolarized after trion decay, i.e. when the trion lifetime is longer than the hole spin relaxation time, then the total spin of the electron ensemble is decreased, in agreement with the simplified Eq. (5) obtained from qualitative arguments. The dependence of the depolarization factor $Q_i^2$ on the control pulse area for different detunings between the trion resonance and the control optical frequencies is shown in Fig. 10. The depolarization efficiency shows Rabi oscillations and is larger for small detunings.

In the case of small control power effective pulse area $\Theta_i \ll 1$, and for negligible detuning between the control pulse and the trion resonant frequency, $y \ll 1$, one can represent $Q_i^2$ in Eq. (12) as

$$Q_i^2 \approx 1 - \frac{(\mu\tau_p)^2}{2}.$$  

(14)

**B. Circularly polarized control**

Now we turn to the case of circularly polarized control pulses. For a $\sigma^+$ polarized control pulse the envelope function

$$f_+(t) = \frac{\mu}{\cosh \left(\frac{\varphi t}{\tau_p}\right)}, \quad f_-(t) = 0,$$

(15)

The time integrated intensities of the circularly polarized pulse $\propto \int_{-\infty}^{\infty} [f_+^2(t) + f_-^2(t)] dt$ and of the linearly polarized pulse, Eq. (11), are the same.

Making use of Ref. [18] we obtain the following expressions which link the spin components before and after control pulse arrival:

$$S_z^{(a)} = \mp \frac{1 - Q_c^2}{4} + \frac{Q_c^2 + 1}{2} S_z^{(b)},$$

(16)

$$S_x^{(a)} = Q_c \cos \Phi_c S_x^{(b)} \pm Q_c \sin \Phi_c S_y^{(b)},$$

(17)

$$S_y^{(a)} = Q_c \cos \Phi_c S_y^{(b)} \mp Q_c \sin \Phi_c S_x^{(b)}.$$  

(18)

Here the upper signs of $\mp$ and $\pm$ correspond to a $\sigma^+$ polarized control and the lower signs to a $\sigma^-$ polarized control. The constant $Q_c$ is given by

$$Q_c^2 = 1 - \frac{\sin^2(\Theta_c/2)}{\cosh^2(\pi y)},$$

where $\Theta_c = \sqrt{2}\Theta_i = 2\mu\tau_p$. For small pulse areas $\Theta_c \ll 1$ and $y \ll 1$

$$Q_c^2 \approx 1 - (\mu\tau_p)^2.$$  

(19)

The modification of the spin $z$ component by a $\sigma^+$ control pulse for different pump pulse areas are shown in Fig. 11. Each curve shows the control pulse area dependence for a fixed pump pulse area $\Theta_0 = 2\mu_0\tau_p$, where $\mu_0$ is the amplitude of the pump pulse envelope as defined in Eq. (15). Rabi oscillations with period $2\pi$ are clearly seen. Here we assumed that the control pulse arrives at $\varphi = 0$, i.e. in the same phase as the pump pulse. Note, that a $\sigma^+$ polarized pump results in an electron spin $z$-projection $S_z < 0$, and corresponds to positive values of the measured Kerr rotation signal, $K(t)$. For convenient comparison of the theoretical and experimental results we invert the direction of the axis of the “spin $z$ component” in the theoretical figures. Here and below the
electron spin dephasing is completely neglected in the calculations. The modification of the spin component $S_z^{(a)}$ comprises both additive and non-additive contributions. Interestingly, for co-polarized pump and control (solid lines) the modification is weaker compared with the cross polarized configuration (dashed lines). This is because the absolute spin value $|S_z|$ is limited by $1/2$ and when the spin projection is closer to $-1/2$ (pump and control are co-$\sigma^+$ polarized) the effect of the control pulse is weaker.

Figure 11 shows the time dependencies of the spin $z$ component calculated for different moments of control pulse arrival, i.e. for different phases of the electron spin precession generated by the pump: $\varphi = 0, \pi, \text{ and } 3\pi/2$. The pump and control pulses are co-circularly polarized. The different curves in each panel correspond to different areas of the control pulse.

One can see from Eq. (19) that there are two contributions to the spin $z$ component of an electron after circularly polarized control pulse arrival. The first contribution is an additive one: it changes its sign upon reversal of the circular polarization of the control pulse and it does not depend on the spin state before control pulse arrival. For weak control power, $\mu\tau_p \ll 1$, and negligible detuning, $y \ll 1$, the additive part to $S_z^{(a)}$ is given by, see Eq. (19)

$$\frac{1}{4} - \frac{Q_c^2}{4} \approx \frac{(\mu\tau_p)^2}{4}. \quad (20)$$

This additive contribution equals exactly the spin $z$ component created by a pump pulse of the same power.

Another contribution to the electron spin after control pulse action is a non-additive one. It can be interpreted as a transformation of the electron spin by the control pulse. This contribution is given by

$$\frac{Q_c^2 + 1}{2} S_z^{(b)} \approx \left[ 1 - \frac{(\mu\tau_p)^2}{2} \right] S_z^{(b)} \quad (21)$$

where the last approximate equality holds for weak control power and small detuning. This non-additive contribution is independent of the circular polarization sign and always decreases the $z$ component of electron spin.
The comparison of Eq. (21) with Eqs. (13) and (14) shows that for weak control powers the depolarization of the electron spin z component by circularly and linearly polarized light is the same.

The in-plane spin components are also affected by the circularly and linearly polarized control pulses. The absolute value of the in-plane spin projection $S_\parallel = \sqrt{S_x^2 + S_y^2}$ is decreased by the factor $Q_c \approx 1 - (\mu \tau_p)^2/2$ (the latter equality holds for weak control pulses), similar to the case of a linearly polarized control. In addition, the detuned circularly polarized control pulse rotates the in-plane spin by the angle $\Phi_c$ around the z-axis.

It is noteworthy to analyze the spin beats phase after circularly polarized control arrival at $\varphi = \pi/2$ where the signal amplitude is zero. In order to calculate the spin beats phase we assume that the magnetic field is applied along the x-axis. We neglect the detuning between the control pulse optical frequency and the trion resonance frequency. Hence, the phase shift of the spin beats induced by the control is given by

$$\theta = \arctan \left( \frac{S_x^{(a)}}{S_y^{(a)}} \right), \quad (22)$$

where the spin precession direction was assumed to be clock-wise in the (yz) plane. The dependence of $\theta$ on the control pulse area is shown in Fig. 13 by the solid line. We compare this phase with the results of the simplified additive model, where we assume that the y spin component is conserved and we take into account the additive contribution of Eq. (10). The phase shift in the additive model is

$$\theta' = \arctan \left( \frac{Q_c^2 - 1}{4S_y^{(b)}} \right). \quad (23)$$

This shift is shown by the dashed line in Fig. 13. The qualitative behaviors of the two shifts $\theta$ and $\theta'$ are the same, however, the exact model predicts a stronger phase shift. This results from the suppression of the in-plane components induced by the circularly polarized light.

Figure 14 shows the electron spin z component after control pulse arrival, calculated as function of control pulse area for two pump pulse areas and for co- and cross-polarized configurations. We assumed that the control pulse arrives at phase $\varphi = 0$ of the spin beats. For a weak pump pulse ($\Theta_0 = \pi/10$) the additive contribution by the control is dominant. The modification of the electron spin component is almost the same in the co- and cross-polarized configurations as it mostly scales with control power. The maximum absolute value of the electron spin projection in this case is close to 0.25, in agreement with Eq. (16) for $S_y^{(b)} \ll 1$.

The case of a strong pump pulse, $\Theta_0 = \pi$, is different. Figure 14 shows a strong asymmetry for the induced polarizations in the co- and cross-polarized configurations. In the cross-polarized case the change of the spin z component is about the same as for a weaker pump. In the co-polarized configuration the control pulse effect is much weaker. This is because the electron spin coherence generated by the pump pulse is partially suppressed by the control pulse. For this configuration the maximum absolute value of the electron spin z component is $1/4 + 1/8 = 0.375$ according to Eq. (16).

Let us also analyze the non-additive effect by the control pulse for the case when it arrives exactly in the maximum or minimum of the spin beats ($\varphi = 0$ or $\pi$), i.e., when the in-plane spin components before control pulse arrival are zero $S_x^{(b)} = S_y^{(b)} = 0$. In this case the electron spin z component is simply suppressed by the
non-additive contribution, in agreement with Eqs. (24) and (21). The efficiency of the spin depolarization is illustrated in Fig. 15. For small pulse areas indeed the depolarization is the same for the linearly and the circularly polarized control. Rabi oscillations are seen with period $2\pi$ for the circularly polarized control and with period $2\sqrt{2}\pi$ for the linearly polarized control. For a circularly polarized control the depolarization is weaker and not complete: one can suppress the spin polarization by no more than a factor of 2, while complete depolarization is possible by linearly polarized light. Note, that complete depolarization is possible for any arrival phase of the control pulse in case of linear polarization.

It is worth to mention that the degree of spin suppression by circularly polarized light is model sensitive. In the following subsection IV.C we demonstrate that the extension of the model to account for the trion excited states could result in stronger spin suppression by the circularly polarized control.

C. Effects of very strong circularly polarized control pulses

Here we analyze briefly the effect of circularly polarized control pulses of very high intensity. We have seen that the model description in terms of a two-level model gives a complete depolarization of the electron spin by linearly polarized light and partial (by a factor of 2, at most) depolarization by circularly polarized light. This is because the transition for a given circular polarization involves just two levels, the ground electron state and an excited (singlet) trion state. Therefore, only one component of the electron spin is pumped into the trion state and becomes subsequently depolarized, while another one is maintained.

There are other possible excited states in the system, e.g. the triplet trion state, which can be populated by polarized light absorption. In the classical approach this state can be considered as an exciton interacting with a resident electron. Due to the electron spin-flip within a triplet trion a singlet trion state can be formed.

To analyze the non-additive effect of a circularly polarized control pulse for the case when the triplet trion/exciton can be photocreased, we denote the probability of singlet trion formation via an exciton [as a result of the following process: electron $-1/2 +$ exciton $(-1/2, 3/2)$, afterwards electron spin-flip and formation of $(-1/2, 1/2, 3/2)$ or $(-1/2, 1/2, -3/2)$ trion] by $W$ and the probability of direct singlet trion formation $[1/2$ electron + photocreased exciton $(-1/2, 3/2)$ yields $(-1/2, 1/2, 3/2)$ trion] as $W'$. Let us do the analysis for the experimental scenario of Sec. III.E where the pump polarization is assumed to be modulated while the control is always $\sigma^+$ polarized. If the electron spin before control arrival is $1/2$ the electron spin after trion recombination is $(1 - W)/2$, because in this case direct singlet trion formation occurs. If the electron spin after control arrival is $-1/2$ then its spin after trion recombination is $-1 + W)/2$, since formation of a triplet trion/exciton is required. The detected signal is suppressed compared to the case without control by the factor

$$S_z^{(a)} = \left(1 - \frac{W + W'}{2}\right) S_z^{(b)}. \quad (24)$$

At high pump powers both $W$ and $W'$ approach unity (see Ref. 10) and the spin after control is completely erased. Clearly, $W$ approaches 1 faster since no electron spin-flip is needed. Therefore one can expect a kind of “two-stage” behavior of suppression: first the spin is suppressed down to the level $(1 - W)/2$ of its value before control pulse arrival, and further increase of control power yields complete suppression.

This process can be described quantum mechanically by extending the wave function $\Psi$, Eq. 4, to allow for the two trion states with total spin projection $\pm1/2$, formed by two spin down electrons and a $3/2$ hole or two spin up electrons and a $-3/2$ hole. For a $\sigma^+$ control pulse the electron $-1/2$ is excited into a $1/2$ triplet trion, and, following 18 we obtain

$$\psi_{-1/2}^{(+\infty)} = \tilde{Q}_e \exp(i\tilde{\Phi}_e) \psi_{-1/2}^{(-\infty)},$$
$$\psi_{1/2}^{(+\infty)} = Q_e \exp(i\Phi_e) \psi_{1/2}^{(-\infty)}.$$

Note that the constants $Q_e$ and $\tilde{Q}_e$ (as well as $\Phi_e$ and $\tilde{\Phi}_e$) are different because the triplet trion is usually shifted.

FIG. 15: (Color online) Suppression of the spin $z$ component by circularly (blue solid) and linearly (red solid) polarized control pulses as function of control pulse area. Thin solid line (black) demonstrates spin suppression by a circularly polarized pulse for the case when both singlet and triplet trion transitions are excited with the same probability, Eq. (20) with $\tilde{Q}_e = Q_e$. The detuning between the quantum dot trion resonance and the control optical frequency is zero. Dashed red curve gives the small amplitude asymptotics.
in energy as compared with the singlet one. If we assume, that after trion recombination the electron is left behind unpolarized, then its spin $z$-component is given by

$$S_z^{(a)} = -\frac{\hat{Q}_c^2 - Q_c^2}{4} + \frac{\hat{Q}_c^2 + Q_c^2}{2}S_z^{(b)},$$

Eq. (25) clearly shows that there are both additive and non-additive contributions to the electron spin $z$ component. The non-additive contribution is

$$S_z^{(a)} = \frac{\hat{Q}_c^2 + Q_c^2}{2}S_z^{(b)}.$$ (26)

One sees that excitation of the triplet trion state results in additional suppression of the electron spin polarization.

We note that the probability of singlet trion formation by a short pulse is $1 - Q_c^2$ and the probability of triplet trion formation is $1 - Q_c^2$. Hence, the quantum and classical approaches are equivalent to each other if we take $W = 1 - Q_c^2$ and $\tilde{W} = 1 - Q_c^2$.

It is instructive to consider two limiting cases:

(i) Only the triplet trion is excited ($Q_c \neq 0$, $Q_e = 0$). The non-additive spin suppression is fully described by the theory developed in Secs. IV A and IV B by changing $Q_c \rightarrow \hat{Q}_c$, $\Phi_c \rightarrow \hat{\Phi}_c$ and replacing ± by ± in Eq. (16). Suppression by the circularly polarized light is possible by a factor 2 only, similar to the situation when only the singlet trion is excited.

(ii) The formation probabilities of the singlet and triplet trions are the same, $Q_c^2 = \hat{Q}_c^2$. The spin suppression factor for circularly polarized control is given by $Q_c^2$, see Eq. (25), i.e. complete depolarization is possible, see thin solid curve in Fig. 15. It is remarkable that in this case the depolarization effect by linearly and circularly polarized controls of the same area are identical.

V. DISCUSSION

A. Comparison theory and experiment

So far, we have established experimentally and theoretically that the control pulse has, in general, a two-fold effect on the electron spin coherence in quantum wells. First, a circularly polarized control pulse generates additional spins and results in an additive contribution to the spin beats. Besides, the control pulse effects the spins that are already polarized by the pump pulse, leading to suppression of the pump-induced spin coherence. The latter effect is possible both for circularly and linearly polarized control pulses.

To do a quantitative comparison of the experimental and theoretical results we consider in detail the effect of the spin coherence suppression by linearly and circularly polarized light. Figure 16 shows the suppression efficiency, i.e. the ratio $S_z^{(a)}/S_z^{(b)}$ as function of control pulse power for a linearly polarized control (closed circles) and a circularly polarized control (open circles). We focus on the small control power regime $P_c \leq 5 \text{ W/cm}^2$ illustrated in detail in Fig. 16(b). In this regime the efficiency of suppression increases linearly with increasing control pulse power. Fitting the experimental data by the theoretical model, Eqs. (14) and (21), we obtain a relation between the control pulse power and its area:

$$P_c = C \Theta_l^2,$$ (27)

where $C \approx 0.63 \text{ W/cm}^2$ is the fitting parameter. The theoretical curve corresponding to the limit of $\Theta_l \ll 1$ is shown by the dashed line in Fig. 16.

The solid thick and thin lines show the suppression efficiency as function of control power for linear and circular polarizations of the control pulse, respectively. They were calculated for the whole range of experimentally used
powers by Eqs. (13) and (21), using the link between the control power and its area from Eq. (27), see Fig. 16(a). The theory reproduces the experimental data well for control powers \( P \lesssim 5 \text{ W/cm}^2 \). For higher powers the discrepancy between experiment and theory is large, the reasons for that are discussed in Sec. VII B.

It is worth to note that other experimental data recorded at low pump powers are in good agreement with the theory. Figure 4 shows the normalized amplitude of the Kerr rotation signal measured as function of control power for co- and cross-polarized configurations. The lines in Fig. 4 show the theoretical calculations obtained from Eq. (10) using the relation between the pulse area and control power Eq. (27) with the same value of \( C = 0.63 \text{ W/cm}^2 \) as in Fig. 16. Good agreement between the experimental data and theoretical curves is seen. Figure 4 shows that for co-polarization the amplitude of the signal saturates faster than for cross-polarization. This is reasonable, because the spin projection of a single electron is limited by 1/2. Therefore, in co-polarization the spin should saturate faster because spin with projection of the same sign is added and, therefore, the spin reaches the maximum value faster.

We also address the phase shift of the spin beats \( \theta \) as function of control power, Fig. 5(b). The black circles shows the phases of the Kerr signal after control pulse arrival extracted from the experimental data. The dashed curve was calculated in the additive model by Eq. (28), and the solid curve shows the theoretical result taking into account additive and non-additive effects, Eq. (22). In both calculations the same relation between the pulse area and its power given by Eq. (27) was used. The solid theoretical curve reproduces well the decrease of the phase shift from 0 to \(-\pi/2\) and its saturation, confirming that indeed non-additive effects need to be considered for a comprehensive analysis.

### B. Effects of high control powers

As mentioned above, Fig. 16 shows significant discrepancies between experiment and theory for control powers \( P \gtrsim 5 \text{ W/cm}^2 \). First, Rabi oscillations are not observed experimentally. Second, linearly and circularly polarized control suppress the spin coherence with about same efficiency, while the theory predicts that the suppression for circular polarization should not exceed 50%.

The absence of the Rabi oscillations shows that the quantum model of Ref. 15 used here is not fully applicable for quantum well structures. Indeed, the classical approach developed in Refs. 10, 19 shows that at high pumping powers saturation effects become important. The classical approach to the description of spin coherence generation and the quantum approach of Ref. 15, extended here to allow for linearly polarized control pulse, coincide exactly in the limit of weak pump and control powers 10. With an increase of control power the quantum mechanical approach predicts Rabi oscillations for the control parameters \( Q_l \) and \( Q_c \). The quantum approach is justified for quantum dots where electrons and trions preserve their coherence on the time scale of pump or control pulse. The applicability of the quantum approach for quantum wells is governed by the relation between the pulse duration \( \tau_p \) and the scattering time between different trion states \( \tau_1 \). If \( \tau_p \ll \tau_1 \) the two-level model, which describes electron to trion excitation under light pulse action, is valid. Otherwise, if \( \tau_p \gtrsim \tau_1 \) the trion can scatter to another state during the pulse action and, therefore, the Rabi oscillations become damped.

To illustrate the transition from the quantum to the classical model we performed calculations of the suppression factor \( S_z^{(a)}/S_z^{(b)} = Q_l^2 \) as function of the linearly polarized pulse area \( \Theta_t \), taking into account a finite scattering time between different trion states. We introduced it as a negative imaginary part \(-i/(2\tau_1)\) of the trion resonance frequency, \( \omega_0 \), in Eq. (9). The calculated depolarization factor is shown in Fig. 17. It is seen that the Rabi oscillations become less pronounced with increase of \( \tau_p/\tau_1 \) and eventually disappear for \( \tau_p/\tau_1 \gtrsim 3 \).

![FIG. 17: (Color online) Depolarization factor for a linearly polarized control as function of control pulse area calculated for different ratios of pulse duration \( \tau_p \) and trion scattering time \( \tau_1 \): \( \tau_p/\tau_1 = 0.6, 2, \) and 6.](image-url)
C. Efficiency of electron spin manipulation

It is instructive to analyze the efficiency of spin control by circularly polarized pulses. To this end we plot in Fig. 18 the absolute value of the spin $z$ component change caused by the control pulse, $|S_z^{(a)} - S_z^{(b)}|$, as function of control and pump pulse areas using Eq. (10). We assume that the pump and control pulses are co-polarized and that the control pulse arrives at $\varphi = 0$ of the spin beats. It is clearly seen that the modification of the spin $z$-component is a non-monotonous function of the pump and control pulse areas. The control efficiency depends strongly on the pump area. For instance, if the pump area corresponds to a $\pi$ pulse, $\Theta_0 = \pi$, i.e. the pump effect is maximal, the control effect is reduced as compared with the case of $\Theta_0 = 0$, where the pump is absent. This is a result of the non-additive effect of the control pulse: if there is already some spin polarization, it is then reduced by the non-additive effect. In other words, the electron spin projection is limited by $|S_z| \leq 1/2$, therefore, the larger is the spin created by the pump, the weaker is the effect of the control that can be realized.

![Graph showing the modification of the spin z component as function of control and pump pulse areas. Panel (a) shows 3D plot. Panels (b) and (c) show its cross-sections for $\Theta_c = \pi$ and $\Theta_0 = \pi$, respectively. Calculations performed for co-polarized configuration and $\varphi = 0$.]

For the cross-polarized control and pump configuration (or in the case the co-polarized control arrives at $\varphi = \pi$ of the spin beats) the spin $z$ component modification is stronger. Indeed, the non-additive effect of the control suppresses the spin polarization and the spin coherence added by the control pulse has an inverse sign as compared with the pump-induced one. Therefore, an increase of the control pulse area from 0 to $\pi$ always increases $|S_z^{(a)} - S_z^{(b)}|$ independent of the pump pulse area, contrary to the co-polarized configuration.

VI. CONCLUSIONS

We have demonstrated experimentally the possibility to manipulate the electron spins in quantum wells by means of polarized laser pulses. We have shown that the coherence of resident electrons can be increased or decreased by a circularly polarized control pulse depending on the pump/control delay and the relative polarizations of the pump and control pulses. This additive effect is a result of spin coherence generation by the control pulse which may be added to or subtracted from the pump-induced spin coherence.

Surprisingly, we have also found a non-additive effect of the circularly polarized control pulse. This contribution is experimentally detected by a special modulation protocol where the control pulse is not modulated while the pump pulse is modulated and the Kerr signal is detected by a lock-in technique. The measured signal is decreased by the control pulse and the suppression efficiency is determined by the control pulse power only. It is independent of the circular polarization of the control pulse and the amount of spin coherence induced by the pump.

A similar suppression is observed for linearly polarized control pulses which do not generate any spin coherence in our geometry. The suppression efficiency is the same for linearly and circularly polarized pulses at relatively small control powers.

The experimental findings are well explained by the proposed theoretical model which takes into account the formation of the singlet trion, localized on an imperfection of an $n$-type quantum well, by polarized light. The electron spin left over from the trion after its radiative recombination is depolarized. Since linearly polarized light results in trion formation regardless of the electron spin projection the spin coherence is suppressed. The model describes both the additive and non-additive effects by circularly polarized control pulses.

The developed model can also be applied to describe the electron spin coherence control in quantum dots. Similarly to quantum well systems studied here, both additive and non-additive effects of the control pulse should be manifested in that case. One may also expect the observation of Rabi oscillations of spin suppression for the quantum dot systems since the trion state is much more robust and observations of Rabi oscillations have been reported, e.g., in experiments with optical generation of spin coherence in an ensemble of singly charged (In,Ga)As/GaAs quantum dots [20].

The manifestations of the non-additive effect are related with the considerable spin polarization generated by the pump pulse, and in general, do not require the
trion as an intermediate state in the spin coherence manipulation. The high spin polarization regime can be achieved for the widely studied quantum wells containing a dense electron gas. However, it occurs at much higher excitation densities where other non-linear effects complicate the interpretation of the experimental data. On the contrary, in quantum wells with a low density electron gas as studied here a relatively high spin polarization can be reached already at rather low excitation powers.

Acknowledgements The authors are grateful to E.L. Ivchenko for valuable discussions. The work was supported by the Deutsche Forschungsgemeinschaft, the EU Seventh Framework Programme (Grant No. 237252, Spin-optronics), the Russian Foundation for Basic Research, the Ministry of Science and Higher Education (Poland) through grant N202 054 32/1189 and by the Foundation for Polish Science through subsidy 12/2007. One of the authors (MMG) acknowledges support by the President grant for young scientists and the “Dynasty” Foundation – ICFPM.

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