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Coherence-mediated laser control of exciton and trion spins in CdTe/CdMgTe quantum wells studied by the magneto-optical Kerr effect

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Abstract

Two temporally non-overlapping linearly cross-polarized 140 fs laser pulses are shown to control the spin polarization in a three-level system. Simultaneous excitation of the two excited states triggers quantum beatings originating from the interference of the wavefunctions corresponding to different spin sublevels of the states. Although the beatings are not seen in the spin densities of the excited states they are clearly observed in the magneto-optical Kerr effect. An analytical expression for the description of the beatings is obtained. Experimental results are in good agreement with theoretical predictions and demonstrate the control of beatings with attosecond resolution.

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

1. Introduction

Optical orientation is one of the main approaches for manipulation of spins in solid-state quantum computing and semiconductor spintronics [1, 2]. The phenomenon of optical orientation is based on the absorption of circularly polarized photons and angular momentum transfer from the photons to electrons. Due to the conservation of angular momentum and the spin–orbit interaction, such an excitation results in an effective generation of a nonequilibrium spin polarization [3]. Over the last 20 years the spin dynamics following optical orientation has been intensively studied and is quite well understood [4]. However, on the timescale of optical decoherence (from 20 fs in metals up to 2 ps in semiconductors) the understanding of spin dynamics is much less evolved since such studies are complicated by the effects of optical coherence and quantum interference [5]. This lack of understanding prohibits optical control of spins at the ultrafast timescale.

Indeed, on such an ultrashort timescale even two temporally non-overlapping linearly cross-polarized pulses, which do not carry any angular momentum, can still effectively generate a spin polarization of the excited state [6, 7]. This becomes possible due to the effects of optical coherence, when a coherent superposition of the optical polarizations induced by these two pulses stimulates the required angular momentum transfer within the electronic system [8]. The process can be understood by considering the helicity components of the linearly polarized optical pulses. The first pulse induces a coherent superposition of spin 'up' and spin 'down' states. If the delay between pulses is smaller than the optical decoherence time, the second pulse will constructively interfere with one spin state, while destructive interference will occur for the other spin state, resulting in a spin polarization of the excited state. Such a coherence-mediated optical orientation of spins was demonstrated in [6] for a two-level system using heavy-hole (hh) excitons as an example. The

feasibility of optical orientation of spins with the help of two linearly cross-polarized laser pulses has been demonstrated for a three-level system containing heavy-hole and light-hole (lh) excitons [7]. It has been shown that by changing the delay between the two pump pulses one can control the total spin polarization, allowing us to generate both parallel and antiparallel spin polarizations of the two excited states. Understanding the subsequent spin-dependent dynamics of this three-level system addresses the fundamental issue of optical control of spins at the ultrafast timescale. Moreover, it appears that the coherence-mediated optical orientation by two linearly polarized pulses may provide a unique opportunity to engineer and control the energy distribution of the spin population in a multi-level system. Such engineering and control would require knowledge about the ultrafast laser-induced processes in the spin system on the timescale of decoherence processes. However, even for the case of a three-level system, the spin-dependent dynamics triggered by two linearly cross-polarized laser pulses has not been investigated so far.

Here we report studies of the spin dynamics in a three-level system excited by two 140 fs linearly cross-polarized pulses. We consider two types of three-level systems with excited states constructed of: (a) hh- and lh-excitons, and (b) hh-excitons and trions. The trion is a complex consisting of an exciton and an electron from the quantum well structure. Using the magneto-optical Kerr effect as a probe we demonstrate that by changing the delay between two linearly polarized pump pulses, one may obtain full coherent control of the spin polarization of the excited states. It is shown that simultaneous excitation of hh-and lh-excitons, or hh-excitons and trions triggers quantum beatings originating from the interference between the wavefunctions of different spin sublevels of the excited states (for instance, the hh-exciton state with spin 'up' interferes with the lh-exciton state with spin 'up' and the lh-exciton state with spin 'down'). A theoretical analysis of the beatings allows us to obtain an analytical expression for the description of these beatings. Experimental results are in good agreement with the suggested theory and show the possibility to control both amplitude and phase of the beatings with an attosecond manipulation of the phase difference between the two pump pulses. Although these studies were focused on excitons and trions in semiconductors, the analysis was performed in terms of a general three-level system so that the conclusions of our work can be generalized to other cases involving ultrafast laser control of spins [9].

2. Experimental methodology

The studied sample is a CdTe/$Cd_{0.78}Mg_{0.22}Te$ multiple quantum well (MQW) structure grown by molecular beam epitaxy on a (100)-oriented GaAs substrate. It contains five 20 nm thick CdTe quantum wells, separated by 110 nm thick barriers. The sample is nominally undoped. The low concentration of resident electrons $n_e = 1.3 \times 10^{10}$ cm^{-2} in the wells is due to residual n-type doping of barriers. Photoluminescence and reflectivity spectra reveal hh-exciton and trion transitions at the energies $\hbar\omega_{hh} = 1.600$ eV and $\hbar\omega_T = 1.598$ eV, respectively (figure 1(a)) [10]. The lh-exciton transition is shifted 15 meV to higher energies from the hh-exciton ($\hbar\omega_{lh} = 1.615$ eV).

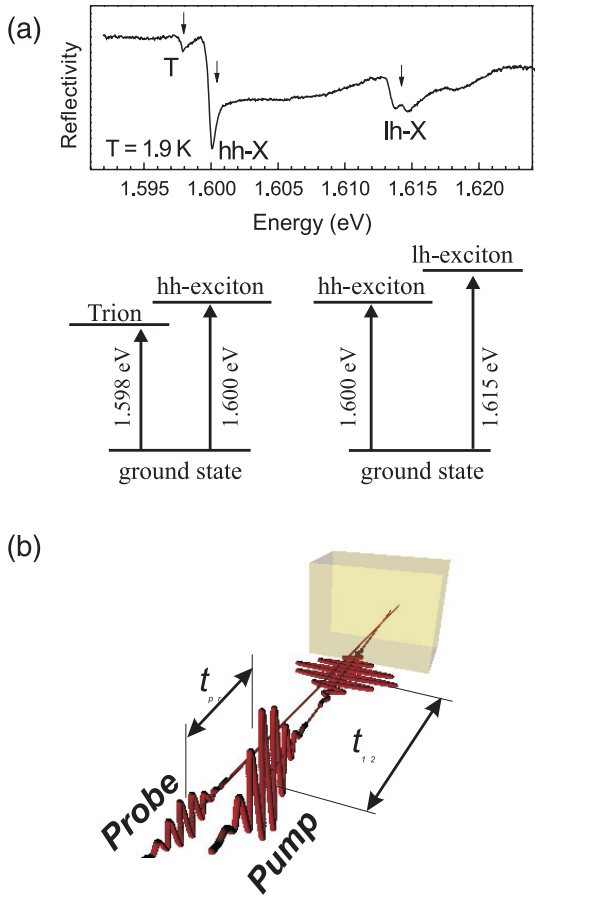

Figure 1. (a) Reflection spectra from CdTe/$Cd_{0.78}Mg_{0.22}Te$ MQW and an energy diagram of the discussed three-level system comprising of hh-exciton (X) and trion (T) as well as hh-exciton and lh-exciton. (b) The experimental scheme with two linearly cross-polarized pump pulses demonstrating the meaning of t_{12} and t_{pr}.

For detection of the laser-induced spin polarization of the excited states we employed the magneto-optical Kerr effect (MOKE). When linear polarized light is reflected from a magnetized sample the polarization rotates over an angle θ

$$i\theta \approx \frac{\chi^+ - \chi^-}{2n(n^2-1)}. \quad (1)$$

Here χ^+ and χ^- are the optical susceptibilities for left- and right-handed circularly polarized light; n is the refraction index. For a three-level system it can be shown that $\chi^+ - \chi^- = a(N^L\uparrow - N^L\downarrow) + b(N^H\uparrow - N^H\downarrow)$, here $N^L\uparrow(\downarrow)$ is the density of the lower excited state with spin up(down), $N^H\uparrow(\downarrow)$ is the density of the higher excited state with spin up(down); a and b are functions of wavelength as well as oscillator strength for the lower and higher-energy transitions, respectively [11]. Therefore the magneto-optical Kerr effect can serve as a measure of total spin polarization of the excited states. It should be noted that the Kerr rotation does not enable us to measure the absolute value of the spin polarization of the system.

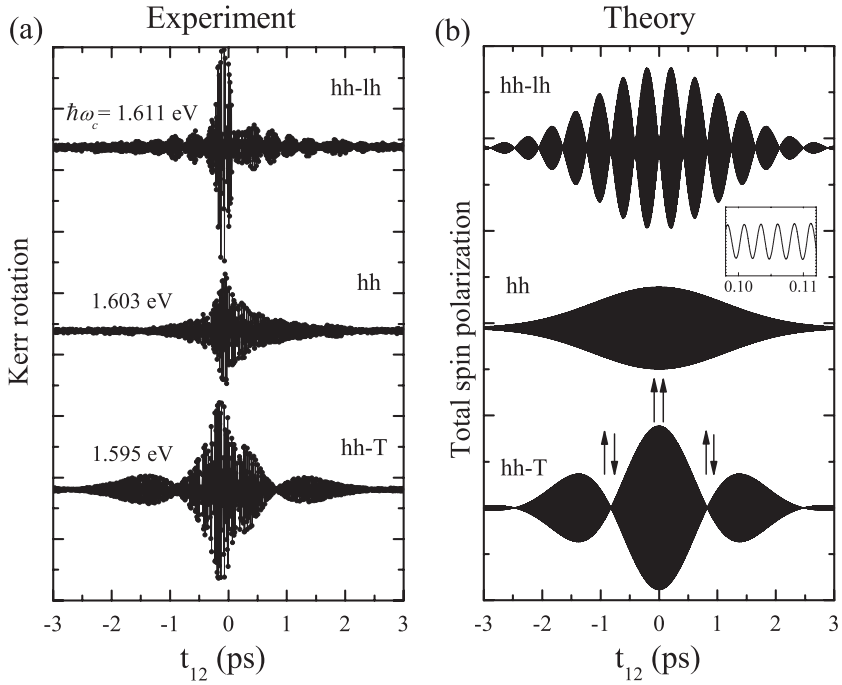

Figure 2. (a) Kerr rotation at probe delay $t_{pr} = 6.7$ ps measured as a function of delay between the two pump pulses t_{12}. The measurements are performed for different central energies $\hbar\omega_c$ of the photons. (b) Simulations of the total spin population of the excited states performed for: (i) simultaneous excitation of hh-exciton and lh-exciton (hh–lh), (ii) hh-exciton only (hh), and (iii) hh-exciton and trion (hh–T). The inset shows the fast oscillations of the total spin population.

For the study of the ultrafast spin dynamics in this three-level system induced by a sequence of two linearly cross-polarized pulses we have employed a pump–pump–probe technique using a mode-locked Ti:sapphire laser that generates pulses with a repetition frequency of 76 MHz. The pulses have a duration of about 140 fs and a spectral width of 10 meV. Both the pulse duration and the spectrum were monitored during the experiment.

The sample was excited by two phase-locked y- and x-polarized pulses separated by an adjustable delay t_{12} (figure 1(b)). For the generation of two phase-locked cross-linearly polarized pump pulses, the laser pulse was sent through an actively stabilized Michelson interferometer. In addition, attosecond modulation of the delay between the two pulses was introduced at $f = 2$ kHz. The laser-induced spin polarization was detected by MOKE of the probe pulses delayed on a time t_{pr} with respect to the x-polarized pump pulses. The polarization rotations of the reflected probe pulses were measured with a balanced photodiode detector. The signal from the detector was measured with a lock-in amplifier, with the 1 kHz modulation f as reference. Using such a phase modulation and lock-in technique we were able to avoid possible artifacts so that the detected signal was the result of the combined action of the two pump pulses. All measurements were performed at a temperature of 10 K.

3. Experimental results

First we demonstrate that a sequence of two linearly polarized pulses can indeed result in spin polarization of the excited states of a three-level system. Figure 2 shows the photo-induced Kerr rotation as a function of the delay between the two pump pulses t_{12}. The delay between the probe and x-polarized pump pulse was fixed at $t_{pr} = 6.7$ ps. At this delay the processes of optical decoherence and thermalization of the exciton and trion sub-systems are expected to be complete, so that the magneto-optical Kerr effect probes the quasi-static laser-induced spin polarization of the excited states [11]. The dependences reveal very fast oscillations of the signal at a frequency close to the frequency of the light. Much slower oscillations with a period of about 0.3 ps are observed on top of the fast ones if the pump's bandwidth covers both hh- and lh-exciton transitions (figure 2(a)) (e.g. the central photon energy is 1.611 eV). If the central frequency of the pump pulses is set to 1.595 eV, so that the spectrum covers hh-exciton and trion transitions, beatings with a longer period of 1.6 ps are seen. If the spectrum covers only one transition, like at $\hbar\omega_c = 1.603$ eV, no beatings are observed. It should be noted that the slight asymmetry in the data is probably due to non-equal absorption for orthogonal polarizations and subsequent non-equal decoherence times. The periods of the observed beatings at central photon energies of 1.595 and 1.611 eV are in good agreement with the energy splitting between the hh- and lh-exciton transitions and the hh-exciton and trion transitions, respectively.

In order to explain such a behavior of the laser-induced spin polarization, we analyze the problem with the help of optical Bloch equations [12]. For simplicity, we neglect processes of spin relaxation, assume the same dephasing time $T_2 = 1.5$ ps and the same oscillator strengths for all the transitions. The pulse durations (140 fs) are taken from the

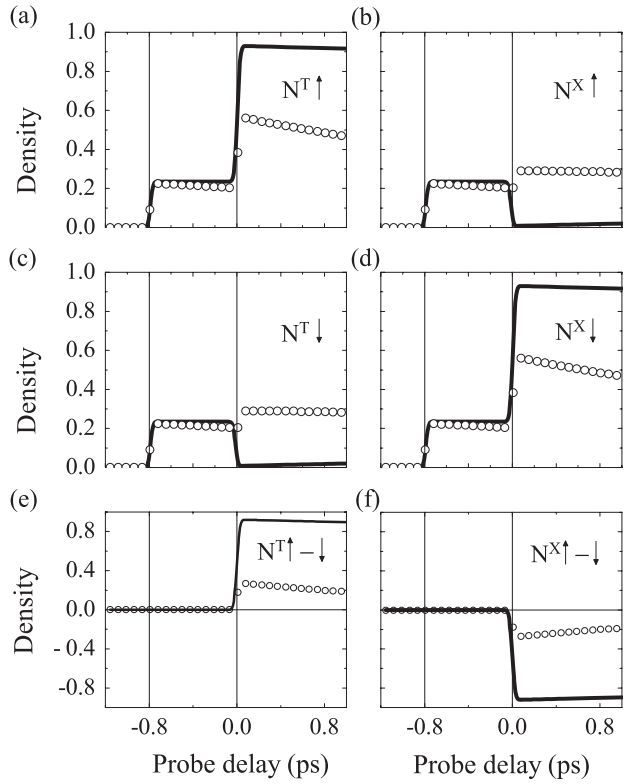

Figure 3. Simulated temporal behavior of the populations generated by two orthogonal linearly polarized pulses arriving at −0.8 and 0 ps. The calculations are performed for a trion with hole-spin 'up' $N^X\uparrow$ (a), and 'down' $N^X\downarrow$ (c), a hh-exciton with hole-spin 'up' $N^T\uparrow$ (b), and 'down' $N^T\downarrow$ (d), $N^T\uparrow - N^T\downarrow$ (e) and $N^X\uparrow - N^X\downarrow$ (f) representing spin polarizations of holes forming excitons and trions, respectively. The calculations are performed assuming negligible spin relaxation so that $T_2 = \infty$ (solid line) or $T_2 = 1.5$ ps (dots).

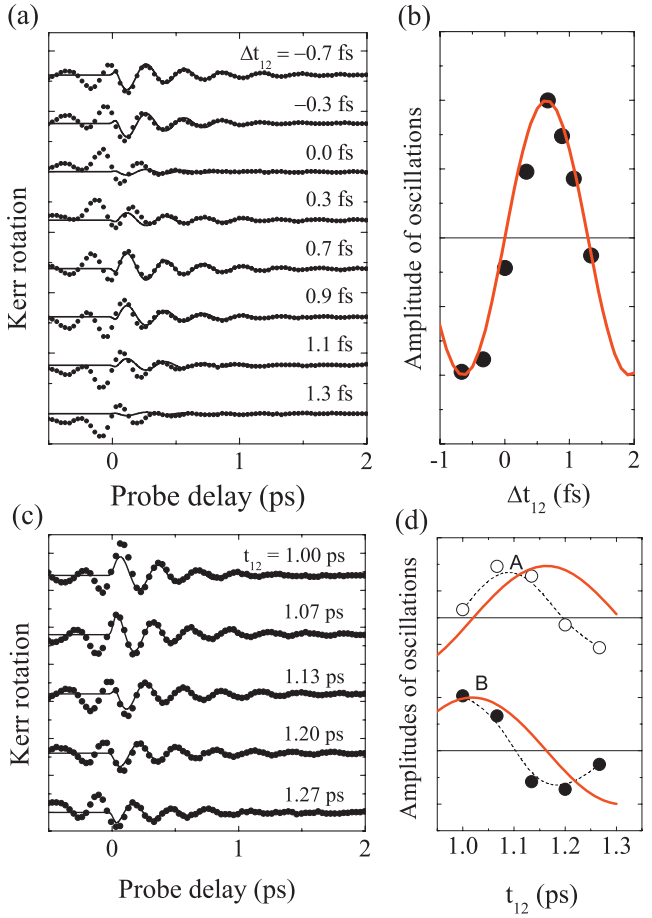

Figure 4. Dynamics of the MOKE signal induced by simultaneous excitation of hh- and lh-excitons measured for different delays between two pump pulses t_{12} for $\hbar\omega_c = 1.611$ eV. Panel (a) shows data where t_{12} is changed in attosecond steps. The solid line represents a fit $f(t_{pr}) = A\cos(\omega t_{pr} + \phi)\exp(-t_{pr}/T_2)$. The best fit was achieved for $\hbar\omega = 13.5$ meV. Panel (b) summarizes the fit showing the dependence of A as a function of Δt_{12}. The solid line is the behavior predicted by equation (7). Panel (c) shows data where t_{12} is changed in femtosecond steps. The solid line represents a fit $f(t_{pr}) = [A\cos(\omega t_{pr}) - B\sin(\omega t_{pr})t]\exp(-t_{pr}/T_2)$. (d) A summary of the results of the fit showing A and B as a function of t_{12}. Solid lines represent the behavior predicted by equation (7), where $\hbar(\omega_{hh} - \omega_{lh}) = 13.5$ meV as deduced from the fit in panel (a). Zero levels of amplitudes in (b) and (d) are given by horizontal lines.

experiment. The total spin polarization at the probe delay $t_{pr} = 6.7$ ps was calculated as a function of t_{12}. The results of the simulations (figure 2(b)) are in good agreement with the experiments. Note that the maxima of the Kerr rotation measured at 6.7 ps after the laser excitation correspond to a parallel configuration of hole-spins forming an exciton and trion (hh- and lh-excitons), while the nulls correspond to antiparallel alignment of the spins. Figure 3 shows the calculated populations of different spin sublevels of the exciton and trion states. The calculations are performed for $T_2 = \infty$ (solid line) and $T_2 = 1.5$ ps (dots). Therefore it can be concluded that resonant excitation of hh- and lh-excitons or hh-excitons and trions by a sequence of two linearly polarized pulses can indeed result in spin polarization of the excited states. At a probe delay of 6.7 ps one may obtain full control over the total spin polarizations of the excited states. It is therefore natural to raise the question about the processes that occur in the spin system between the moment of the laser excitation and 6.7 ps.

To study the initial ultrafast processes in the spin system triggered by two linearly cross-polarized laser pulses, we have measured the magneto-optical Kerr rotation as a function of delay t_{pr} between the x-polarized pump and the probe pulses. Figure 4 shows the Kerr rotation for simultaneous excitation of the hh- and lh-excitons (central photon energy is 1.611 eV). The delay between the two pump pulses was set to $t_{12} = 1.13$ ps $+ \Delta t_{12}$, where Δt_{12} represented the DC attosecond changes of the delay. One can clearly distinguish oscillations with a period of about 0.3 ps.[5] This period is in excellent agreement with the energy difference between the hh- and lh-exciton states. At the same time, no oscillations of the spin densities of the excited state are observed in the simulations (see figure 3).

[5] Note that the beatings are also visible at the negative delays. Indeed if the separation between the pump and probe pulses is shorter than the time of optical decoherence, the pump can still affect the probe even if the pump arrives later than the probe. Such a phenomenon has been seen previously in time-resolved experiments [13, 14].

4. Discussion and theoretical modeling

To understand these periodic variations in the MOKE signal one should realize that the excited state of our system can be described as a coherent superposition of four states. In the case of the hh-/lh-exciton system the wavefunction of the excited state is $\Psi = C_1 e^{iE_1 t}\Psi_1^{(0)} + C_2 e^{iE_2 t}\Psi_2^{(0)} + C_3 e^{iE_3 t}\Psi_3^{(0)} + C_4 e^{iE_4 t}\Psi_4^{(0)}$, where the complex amplitudes C_1 and C_2 represent lh-excitons with hole-spins directed 'up' and 'down', respectively, C_3 and C_4 represent hh-excitons with hole-spins directed 'up' and 'down', respectively. In the absence of a magnetic field the spin levels are degenerate, i.e. $E_1 = E_2 = \omega_{lh}$ and $E_3 = E_4 = \omega_{hh}$. Describing the laser-induced magneto-optical Kerr effect in terms of a nonlinear optical polarization of third order [15, 11], neglecting all relaxation processes in the optically excited semiconductor, and considering the pump and probe pulses as delta functions, we obtain an analytical expression for the magneto-optical Kerr effect (θ)

$$\theta \approx d^2(|C_1|^2 - |C_2|^2) + d^2 r^2(|C_4|^2 - |C_3|^2) + d^2 r\{(C_1 C_4^* - C_2 C_3^*)e^{i(\omega_{lh}-\omega_{hh})t_{pr}} + \text{c.c.}\}. \quad (2)$$

Here d is the parameter proportional to the dipole matrix element, and r is a coefficient that accommodates the difference between the oscillator strengths of the excited transitions. Note that the first two terms are proportional to the total spin polarization of the excited states and do not reveal any time dependence. The third term originates from the quantum coherence between the two excited states, and gives rise to quantum beating in the spin polarization between hh- and lh-exciton states. The beating shows up in the magneto-optical Kerr effect as an oscillation at the frequency given by the splitting between the excited states $\omega_{lh} - \omega_{hh}$.

If one considers excitation with two linearly cross-polarized pulses separated by a delay t_{12}, the complex amplitudes for hh-exciton and lh-exciton spin states become:

$$C_1 = d_{hh}\epsilon(\omega_0 - \omega_{hh})[1 - ie^{i\omega_{hh}t_{12}}] \quad (3)$$

$$C_2 = d_{hh}\epsilon(\omega_0 - \omega_{hh})[1 + ie^{i\omega_{hh}t_{12}}] \quad (4)$$

$$C_3 = d_{lh}\epsilon(\omega_0 - \omega_{lh})[1 + ie^{i\omega_{lh}t_{12}}] \quad (5)$$

$$C_4 = d_{lh}\epsilon(\omega_0 - \omega_{lh})[1 - ie^{i\omega_{lh}t_{12}}] \quad (6)$$

where ϵ is the spectral component of the laser field at the hh-exciton and lh-exciton resonances. It should be noted that for sufficiently spectrally broad laser pulses this spectral component has the same amplitude for both excited transitions. With these obtained complex amplitudes for these spin states, the coherent and incoherent contributions in the case of two-pulse excitation can be analyzed in terms of the delay time between pump pulses (t_{12}). The equation for this 'coherent' term in the Kerr effect (θ_{coh}) for an hh-/lh-exciton system becomes:

$$\theta_{coh} \approx r\epsilon(\omega_c - \omega_{hh})\epsilon(\omega_c - \omega_{lh}) \sin\left(\frac{\omega_{hh} + \omega_{lh}}{2}t_{12}\right) \times \left[\cos\left(\frac{\omega_{hh} - \omega_{lh}}{2}t_{12}\right)\cos((\omega_{hh} - \omega_{lh})t_{pr}) - \sin\left(\frac{\omega_{hh} - \omega_{lh}}{2}t_{12}\right)\sin((\omega_{hh} - \omega_{lh})t_{pr})\right] \quad (7)$$

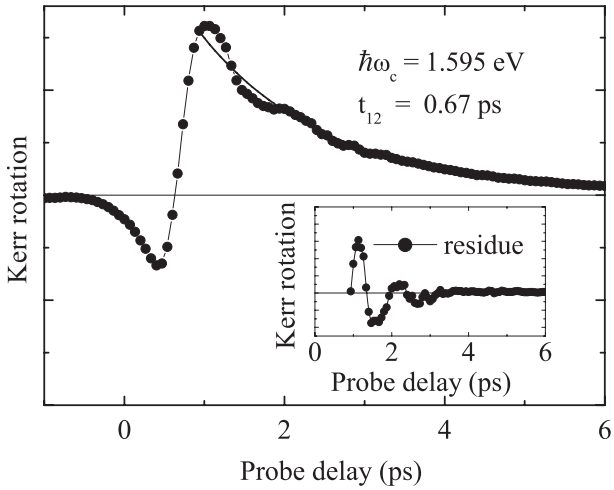

Figure 5. Dynamics of the magneto-optical Kerr effect signal induced by simultaneous excitation of an hh-exciton and trion measured for a delay between two pump pulses $t_{12} = 0.67$ ps. The solid line shows the fit by an exponentially decaying function. On the top of the exponential decay one can distinguish oscillations shown in the inset after subtracting the decaying component shown by solid line. These oscillations correspond to quantum beatings between hh-exciton and trion states with opposite spin orientations.

where ϵ is the spectral component of the laser pulse at the hh-and lh-exciton resonances. From this equation it follows that beating as a function of probe delay (t_{pr}) will be observed with a frequency ($\omega_{hh} - \omega_{lh}$), which equals a period of 0.3 ps measured in the experiments. The amplitude of the beating is proportional to $\sin[(\omega_{hh} + \omega_{lh})t_{12}/2]$ which is in good agreement with the experiment (figure 4). Finally, one can see that, if t_{12} is changed in picosecond steps, the amplitude and the phase of the oscillations changes, closely following the behavior predicted by equation (7). The difference between theory and experiment is seen as a phase shift of about 200 fs, which can be easily understood by the fact that the theory considers the laser pulses as delta functions. All the features clearly demonstrate that the oscillations in the magneto-optical signal from a hh-/lh-exciton system excited by a sequence of two linearly cross-polarized pulses is related to quantum beating between spin sublevels of the excited states corresponding to hh-excitons and lh-excitons, respectively. Varying the delay between the two pump pulses t_{12} we could change the amplitude and the phase of the beatings and thus coherently control the laser-induced spin dynamics at an attosecond timescale.

Finally, it should be mentioned that the beatings between spin densities of the excited states are also observed in the hh-exciton and trion system. In this case the spin density shows beating, with a period of 1.6 ps (figure 5), which corresponds to the energy splitting between these states. Although quantum beatings between exciton and trion states have been observed before [16], to the best of our knowledge quantum beatings between spin sublevels of the excited states have not been reported until now. In contrast to earlier experiments with parallel polarized pump pulses, two perpendicularly polarized pulses used in our experiment do not interfere directly even if

they overlap in time [7]. For this reason, the delay between the two orthogonal pump pulses does not influence exciton and trion densities while playing a dominant role in the formation of spin polarization of the excited states.

5. Conclusions

In conclusion, the optical orientation of spins by two 140 fs linearly polarized pulses and the subsequent spin dynamics in a three-level system has been analyzed for the examples of hh- and lh-excitons as well as the hh-exciton and trion. Using the magneto-optical Kerr effect as a probe, we demonstrate that changing the delay between the two linearly polarized pump pulses, one may obtain full coherent control of the spin polarizations of the excited states. It is shown that simultaneous excitation of hh-and lh-excitons or hh-excitons and trions triggers quantum beatings originating from the interference between the wavefunctions of spin sublevels of different excited states. These beatings are not seen in the spin polarization of the excited states while they are clearly resolved in the magneto-optical Kerr effect. A theoretical analysis of the beatings allowed us to obtain an analytical expression for the description of these beatings. The experimental and theoretical studies as demonstrated show the possibility to control spin-dependent dynamics with unprecedented 'attosecond' resolution.

Acknowledgments

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