Photoinduced magneto-optical Kerr effect and ultrafast spin dynamics in CdTe/CdMgTe quantum wells during excitation by shaped laser pulses

J. H. Versluis,[1] A. V. Kimel,[1] V. N. Gridnev,[2] D. R. Yakovlev,[2,3] G. Karczewski,[4] T. Wojtowicz,[4] J. Kossut,[4] A. Kirilyuk,[1] and Th. Rasing[1]

[1]*Institute for Molecules and Materials, Radboud University Nijmegen, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands*

[2]*Ioffe Physical Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia*

[3]*Experimentelle Physik II, Technische Universität Dortmund, D-44221 Dortmund, Germany*

[4]*Institute of Physics, Polish Academy of Sciences, PL-02668 Warsaw, Poland*

(Received 18 March 2009; revised manuscript received 24 November 2009; published 18 December 2009)

The photoinduced magneto-optical Kerr effect during the excitation by picosecond laser pulses is investigated in $CdTe/Cd_{0.78}Mg_{0.22}Te$ quantum wells in the spectral range of heavy-hole exciton and trion transitions. Using polarization pulse shaping we were able to generate circularly polarized laser fields with an independent control of temporal and spectral resolution. Changing the frequency and temporal position of the circularly polarized field we show that "off-resonant" optical transitions have a considerable influence on the ultrafast dynamics of the magneto-optical Kerr effect. An excitation of a heavy-hole-exciton with an off-resonant circularly polarized field results in a spin polarization of the electrons in the excited state. Such an "off-resonantly" excited spin population is present only during the action of the laser pulse, but quantum interference of off-resonant and "on-resonant" components leads to beats of the spin population of the excited state and similar beats of the magneto-optical signal with frequency and amplitude determined by the shape of the pump pulse.

DOI: 10.1103/PhysRevB.80.235326 PACS number(s): 78.47.J−, 42.50.Md, 72.25.Fe, 73.21.Fg

I. INTRODUCTION

The interaction of circularly polarized light with electronic states influenced by spin-orbit coupling provides a mechanism for spin manipulation in nonmagnetic and magnetic solids.[1–7] During the last decades the spin dynamics triggered by picosecond or subpicosecond circularly polarized laser excitation has been subject of intense research interest.[8–10] A pump-and-probe method, where the laser-induced nonequilibrium spin polarization was probed using the magneto-optical Kerr effect, was shown to be an effective tool for experimental studies of ultrafast spin dynamics. As a result, a considerable progress has been achieved in understanding the magneto-optical signal and spin dynamics that follows the ultrashort laser excitation.[8–10] Nevertheless, there is still no clear picture of the processes that define the photoinduced magneto-optical Kerr effect *during* the action of the laser pulses.

Indeed, numerous all optical pump-and-probe experiments reveal strong magneto-optical signals during the action of a circularly polarized laser pulse. On the one hand, these signals are interpreted as an optical orientation of spins followed by ultrafast spin scattering via intrinsic spin-flip mechanisms on a time scale much shorter than the duration of the pump pulse.[11–13] On the other hand, the ultrafast laser-induced processes that occur during the pump pulse can be described in terms of orientation of electronic orbitals which gets lost due to optical decoherence.[14,15] However, none of these interpretations really takes into account the complex nature of this problem. In particular, during the excitation by an ultrashort laser pulse, which is spectrally broadened, the effects of optical coherence and quantum interference are very important.[16] The influence of these effects on the photoinduced magneto-optical signals, optical orientation of spins and their subsequent dynamics is largely unknown. This is mainly because for a long time there was no tool to control these effects in a medium. Recently, however, this problem has been elegantly solved using shaping of ultrashort laser pulses.[17,18]

Here we aim to understand the processes that define the ultrafast dynamics of the photoinduced magneto-optical Kerr effect during excitation by a short laser pulse. For these systematic studies we employ a polarization pulse shaping method[17,18] and use the pulse shape as an experimental variable. The studies were performed on CdTe/(Cd,Mg)Te semiconductor quantum wells in the range of the trion and heavy-hole (hh) exciton transitions. We demonstrate that the ultrafast dynamics of the magneto-optical Kerr effect cannot be explained by just ultrafast spin scattering via intrinsic spin-flip mechanisms as suggested in.[11–13] Instead, an "off-resonant" (virtual) optical excitation of an exciton transition appears to have a considerable influence on the magneto-optical signal during excitation by a short laser pulse.[20] The off-resonant (virtual) optical excitation is the excitation by a laser pulse having a circularly polarized component either above or below the exciton transition. Such an excitation generates a spin polarization of electrons in the excited state which is present only during the action of the pump pulse. Due to the effects of quantum interference a simultaneous excitation of "on-resonant" and off-resonant components is shown to lead to temporal oscillations of the laser-induced spin polarization of the excited state in a two-level system (particularly the hh exciton), with a frequency and amplitude that are determined by the detuning of the off-resonant frequency from the resonant one. The paper is organized as follows. Section II describes the properties of a $CdTe/Cd_{0.78}Mg_{0.22}Te$ semiconductor quantum well, where the heavy-hole-exciton transition was used as a model system. In the same section we describe the used method of polarization pulse shaping and setup for time-resolved mea-

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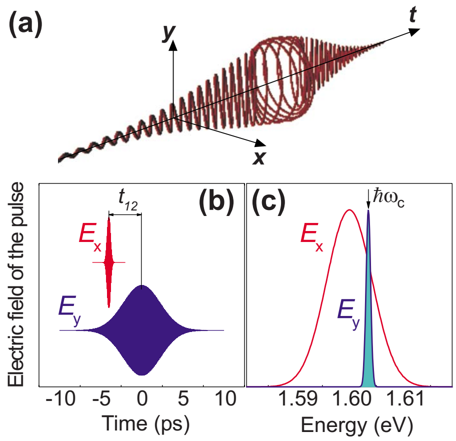

FIG. 1. (Color online) (a) Schematic representation of the shaped laser pulse. (b) Temporal and (c) spectral profiles of the x- and y-polarized components of the laser pulse. It is seen that a circularly polarized state for the shaped pulse is generated within the narrow spectral range of the y-polarized pulse and the ultrashort time window of the x-polarized pulse. It allows independent control of spectral and temporal resolution in optical orientation of spins.

surements of the photoinduced Kerr effect. Section III summarizes the experimental results on the transient magneto-optical Kerr effect induced by polarization shaped laser pulses and theoretical modeling of the latter. Comparing the theory and the experiment we draw the conclusions summarized in Sec. IV.

II. EXPERIMENTAL METHODOLOGY

The sample used for this study contains $CdTe/Cd_{0.78}Mg_{0.22}Te$ modulation-doped multiple quantum wells (MQW), for sample details see Ref. 10. The photoluminescence spectrum exhibits hh exciton (X) and trion (T) energies at 1.600 and 1.598 eV, respectively. Although the selection rules for the exciton and trion are the same, the resonance strength of the trion is almost two times weaker.[24] Sample temperature during experiments was 10 K.

To obtain polarization pulse shaping, that is, spectrally broad laser pulses with polarization being a function of photon energy,[17,18] we used the interference of two linearly polarized Fourier limited laser pulses with orthogonal polarizations. The x-polarized laser pulse was temporally short [full width at half maximum (FWHM)=140 fs] and spectrally broad ($\Delta\omega=10$ meV, $\omega=1.600$ eV), while the y-polarized pulse was temporally broad (FWHM=5 ps) and spectrally narrow ($\Delta\omega=0.3$ meV) with a central frequency ω_c. A coherent superposition of the x and y pulses is a polarization shaped pulse. The phase between the two interfering pulses was set to $+\pi/2$ or to $-\pi/2$ [see Fig. 1(a)]. Circularly polarized fields created in such a fashion are expected to be generated only within the duration of the temporally short x pulse and in the spectral range of the spectrally narrow y pulse. In this way, *spectral and temporal resolution of optical spin orientation can be controlled independently*. This is in strong contrast with previous experiments on optical orientation, which employed transform limited circularly polarized laser pulses. It provides extra degrees of freedom for our study allowing new insights in the problem of ultrafast dynamics of the photoinduced Kerr effect.

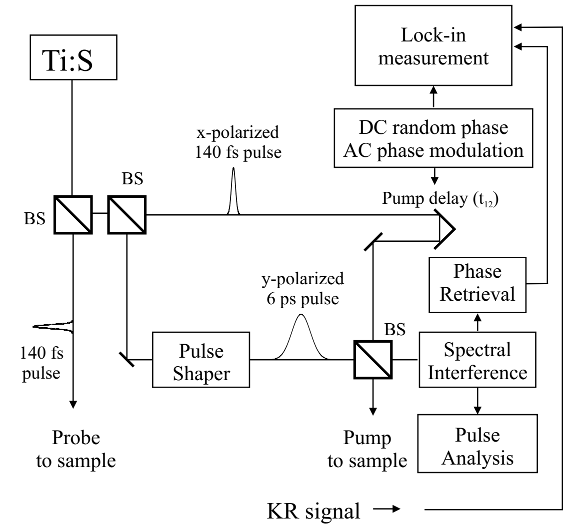

FIG. 2. Schematic picture of pump part of the pump-probe setup. The unshaped pulse is shifted in time with respect to the shaped pulse by a delay stage. Modulation of the phase allows for a spin-sensitive lock-in measurement technique laser-induced spin densities. The random phase is measured by taking the spectral interference between the shaped and the unshaped pulse (reference), which also enables us to do a direct pulse analysis of both the amplitude and phase of the shaped laser pulse. The induced spin dynamics is measured by delaying the probe pulse with respect of the pump pulse.

Experimentally, the photoinduced magneto-optical Kerr effect was measured in a pump-and-probe configuration using a mode-locked Ti:sapphire laser that generated pulses with a repetition frequency of 76 MHz.[14] The induced spin polarization was detected as a function of the delay between the x-polarized part of the pump pulse and the probe. The polarization rotation of this reflected probe pulse (140 fs, $\omega=1.600$ eV) was measured by a balanced photodiode scheme. The measurements were performed for different delays t_{12} and central frequencies of the y-polarized pulse ω_c (see Fig. 2). It must be noted that working with these phase locked pulses one should keep in mind that an attosecond small drift of the delay t_{12} may seriously affect the results of a measurement. In order to solve this problem we have created a random phase modulation between the x- and y-polarized parts of the pump pulse on the top of the fixed offset t_{12}, in the range between $-\pi/2$ and $+\pi/2$. The envelopes of the curves measured in such a way correspond to the spin dynamics induced by the complex pump pulse, where a small part of the spectrum centered at ω_c is circularly polarized, while the rest of the spectrum has the linear x polarization. During the experiments, the spectral properties of the pulse due to pulse shaping were analyzed by taking the spectral interference between the shaped and unshaped laser pulse (acting as a reference pulse). This type of pulse analysis gives direct amplitude and phase information of the spectral components of the shaped pulse. Such an essential measurement allows us to record the phase difference between

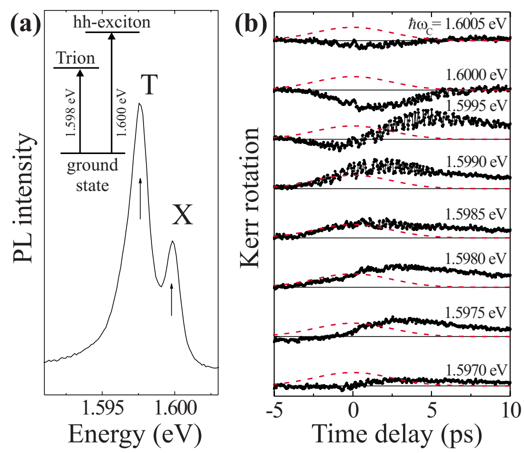

FIG. 3. (Color online) (a) Photoluminescence spectra of $CdTe/Cd_{0.78}Mg_{0.22}Te$ MQWs and the energy diagram consisting of the ground state, hh-exciton (X), and trion (T) levels. (b) Dynamics of the magneto-optical Kerr rotation induced by transform limited 5 ps circularly polarized laser pulses with different central energies of the photons. Dashed lines show the pump pulse.

pump pulses during the performed experiments. With this phase information, phase sensitive measurements could be retrieved from the ensemble of randomized data points. Of course, a large amount of data points will have to be recorded to obtain a phase sensitive spin dynamics measurement with an acceptable phase resolution. In addition, we have performed standard time-resolved pump-probe measurements with a single transform limited 5 ps circularly polarized pump and a 140 fs probe pulse, as explained elsewhere.[14]

III. EXPERIMENTAL RESULTS

A. Transient magneto-optical Kerr effect induced by unshaped circularly polarized pulses

Figure 3(b) shows the temporal evolution of the magneto-optical Kerr effect induced by a transform limited circularly polarized 5 ps laser pulse. The central frequency of the pump was varied in the range from 1.5970 to 1.6005 eV. The cross-section of the Kerr signals at 6 ps shows resonances at 1.5995 and 1.5980 eV, corresponding to the exciton and trion transitions. It should be noted that the slight discrepancy between the hh-exciton resonance observed in this experiment and other measurements presented in this manuscript (<0.5 meV) is probably due to pump-induced energy shift of the hh-exciton energy level, which is beyond the scope of this manuscript. The dynamics of the Kerr rotation during excitation by the pump shows strong wavelength dependence. The origin of the changes cannot be revealed on the basis of these experiments. In order to understand the processes that define the ultrafast magneto-optical Kerr effect during the excitation by a short laser pulse we have studied the Kerr effect using the pulse shape as an experimental variable.

B. Transient magneto-optical Kerr effect induced by polarization shaped pulses

First, we excited the semiconductor by the shaped laser pulses with $t_{12}=0$ and variable $\hbar\omega_c$ [Fig. 4(a)]. Only at delays longer than 5 ps the dynamics excited by the shaped pulse agrees with observed dynamics excited by similarly long transform limited circularly polarized pulses. Indeed, taking Figs. 3(b) and 4(a) and making a cross-section at 6 ps, one can see that the spectral dependencies of the signal induced by the transform limited pump and the polarization shaped pump are very similar (both exhibit maxima close to 1.5980 and 1.6000 eV). At delays shorter than 5 ps the observed dynamics is very sensitive to the shape of the pulse and in particular to $\hbar\omega_c$. At $\hbar\omega_c=1.6000$ eV, a fast relaxation with a decay time of 2 ps is observed, while in the range between 1.5970 and 1.5995 eV beatings of the magneto-optical signal can be clearly distinguished. In order to study these beatings in more detail we tuned $\hbar\omega_c$ to the energy of the trion transition 1.5980 eV and varied t_{12}. It is seen from Fig. 5(a) that a change in the delay t_{12} does have a strong effect on the amplitude of the beatings while the period of the beatings is not affected. Note that the beatings are observed only during the action of the pump pulses.

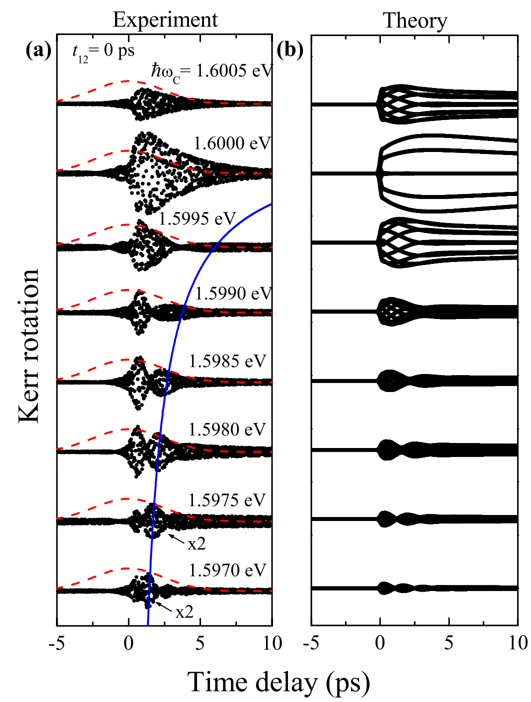

FIG. 4. (Color online) Ultrafast dynamics of the magneto-optical Kerr rotation induced by (a) polarization shaped laser pulses with different $\hbar\omega_c$. The solid line across the graph aims to demonstrate the shift in the beatings when $\hbar\omega_c$ is changed. (b) Simulations of the dynamics for the magneto-optical Kerr effect induced by the polarization shaped laser pulses with $t_{12}=0$ ps and the same $\hbar\omega_c$ as experiments. The duration of the probe and pump pulses is assumed to be equal to those in the experiment, $T_2=2$ ps. To simulate the randomization of the phase between the two pump pulses we have performed the calculations for several phase differences in the range between $-\pi/2$ and $+\pi/2$. The envelopes that enclose these curves are to be compared with those observed in the experiment (a). Dashed lines show the positions of the y-polarized part of the pump pulse.

We have also succeeded to realize direct measurements of the phase difference between the x- and y-polarized parts of the pump pulse so that the spin dynamics that corresponds to

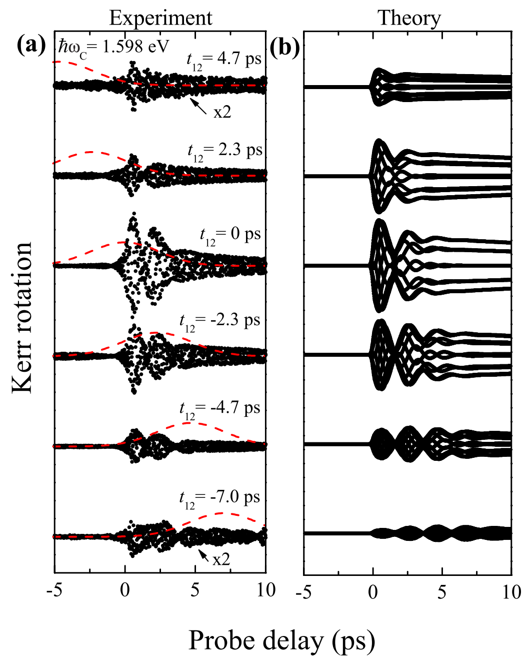

FIG. 5. (Color online) Ultrafast dynamics of the magneto-optical Kerr rotation induced by (a) polarization shaped laser pulses for $\hbar\omega_c=1.5980$ eV and different t_{12}. (b) Simulations performed for the $\hbar\omega_c=1.5980$ eV and the same t_{12} as experiments. To simulate the randomization of the phase between the two pump pulses we have performed the calculations for several phase differences in the range between $-\pi/2$ and $+\pi/2$. The envelopes that enclose these curves are to be compared with those observed in the experiment (a).

a particular phase difference has been retrieved from the ensemble of data points shown in Figs. 4(a) and 5(a). The total range of phases was divided into five regions (Fig. 6). The

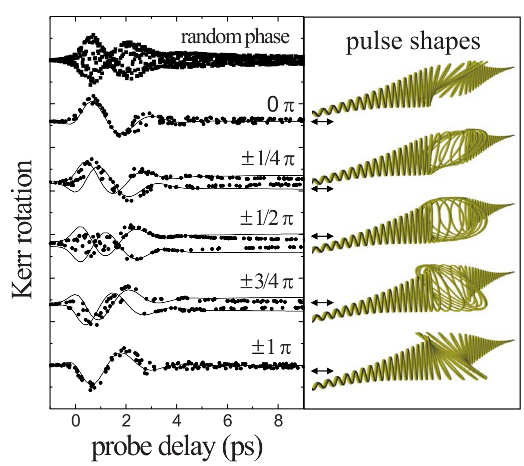

FIG. 6. (Color online) Dynamics of the Kerr rotation as a function of probe delay measured at fixed phase difference between the x- and y-polarized parts of the pump pulse for $\hbar\omega_c=1.598$ meV and $t_{12}=0$ ps. The solid lines are guides to the eye, showing the dynamics for a single pump pulse phase difference. The right panel shows the schematic representation of the pulse shapes corresponding to the given delays between x- and y-polarized parts of the pump pulse.

interference signal was taken as a measure for the absolute value of the phase difference. Note that such a procedure does not allow to extract the sign of the phase. For this reason, if the phase was not an integer number of π, the procedure of retrieval of the spin dynamics at the fixed phase delivered two solutions corresponding to positive and negative phase difference, respectively. The measurements at fixed phase clearly show beatings of the Kerr rotation with a period of about 2.2 ps, corresponding to an energy difference of 2 meV, which matches the detuning from the hh-exciton transition at 1.600 eV (Fig. 6). Note that at times longer than 6 ps the Kerr rotation is the largest for phase differences of $\pi/2$ and $-\pi/2$, showing that a long living spin polarization of the excited state can only be generated by a circularly polarized field. Nevertheless, at the shorter time scale even pulses with phase difference 0 and π, which do not contain circular polarized fields, can trigger spin beatings and the phase of the beatings is directly related to the phase difference between the pump pulses (see Fig. 6).

C. Theoretical modeling and discussion

In order to reveal the nature of the magneto-optical signal during the action of the pump pulse and understand the origin of the observed beatings, we first consider the resonant interaction between a two-level system and a short laser pulse. Neglecting relaxation, first order time-dependent perturbation theory predicts the temporal population of the excited state at the energy $\hbar\omega_0$ to be

$$N(t)\propto\left|\int_{-\infty}^{t}E(t')e^{-i\omega_0 t'}dt'\right|^2, \tag{1}$$

where E is the electric field of light and ω_0 is the resonance frequency of the two-level system.[19] Already from this equation one can see that even off-resonant excitation at a frequency $\omega\neq\omega_0$ may result in a population of the excited state $N(t)\neq 0$, which, however, is present only during the action of the laser pulse. It is easy to show that even in the case of a biharmonic laser excitation at the frequencies $\omega_1=\omega_0$ and $\omega_2=\omega$ one can distinguish between "on-resonantly" and "off-resonantly" excited populations at frequencies 0 and $2(\omega-\omega_0)$, respectively. On top of that, these two contributions may interfere leading to an extra term in the population oscillating at $\omega-\omega_0$. Consequently, one can expect similar beats in the spin population of the excited state that consequently will show up in the magneto-optical Kerr signal.

To confirm the hypothesis of the quantum beats in the spin population of the excited state as the origin of the oscillations seen in the magneto-optical signal we perform simulations of the Kerr effect[14] for the three-level system as explained in Ref. 14. In the simulations the change in the polarization state of the probe pulse upon reflection from the optically excited sample is described by

$$\theta\approx\int_{+\infty}^{-\infty}dtE(t)[P^{++}-P^{--}], \tag{2}$$

where θ is the Kerr rotation. The nonlinear polarizations P^{++} (P^{--}) are a linear function of the probe optical electric field

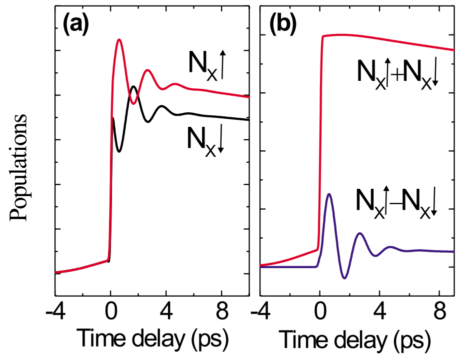

FIG. 7. (Color online) (a) Simulations of the dynamics of the populations of the excited state for two spin sublevels $N_X\uparrow$ and $N_X\downarrow$. (b) Simulations of the dynamics of the total population of the excited state $N_X\uparrow+N_X\downarrow$ and its spin polarization $N_X\uparrow-N_X\downarrow$. The simulations are performed for the case corresponding to pumping with polarization shaped laser pulses with $\hbar\omega_c=1.5980$ eV and $t_{12}=0$.

and a quadratic function of the optical electric field of the right (left)-handed polarized pump pulse.[21] The simulations of the polarizations were based on the optical Bloch equations.[22] To account for the different strengths of the exciton and trion resonances, the exciton was assumed to be four times stronger than the trion. For both transitions the optical decoherence time T_2 was assumed to be 2 ps and relaxation of the pump-induced spin polarization and energy redistribution of the laser-excited electrons were neglected. In order to simulate the randomization of the phase between the x and y polarized parts of the pump pulses we have performed the calculations for several phase differences in the range between $-\pi/2$ and $+\pi/2$. The envelopes that enclose these curves are to be compared with the envelopes observed in the experiment. It is seen from Fig. 4(b) that the simulations performed for different $\hbar\omega_c$ at fixed $t_{12}=0$ ps clearly reproduce the beatings observed in the experiment of Fig. 4(a). The frequency of the experimental beatings decreases with decrease of the difference $\omega_c-\omega_{hh}$ (ω_{hh} is the hh-exciton frequency) and disappears once $\omega_c=\omega_{hh}$. Closer inspection shows that the period of the beatings is proportional to $2\pi/(\omega_c-\omega_{hh})$, as guided by the solid curved line in Figs. 4(a). The relaxation of the experimental signal observed at $\hbar\omega_c=\hbar\omega_{hh}$ was not reproduced in the calculations and thus is likely related to neglected effects of relaxation of spins and charges.

The simulations performed for a fixed $\hbar\omega_c=1.5980$ eV and different t_{12} are also in good agreement with the experiment [Fig. 5(a)]. Similar to the experiment, the beatings are observed only during the action of the pump and their amplitude decreases with increasing t_{12}. Note that due to the smaller trion oscillator strength, we can only see beatings of the exciton spin population. However, the calculations also show that if the trion transition is neglected completely in the present realization of the measurement procedure the beatings will averaged out and will not be seen. On-resonant excitation of the trion is crucial for the outcome of our experiment.

In addition to the calculated Kerr rotations, these simulations provide direct information about the induced populations of the excited states with spin "up" and "down." The total population of the excited state and its spin polarization are shown in Fig. 7 as a function of time. We have chosen a pump pulse with $\hbar\omega_c=1.598$ eV, ($t_{12}=0$), and a phase difference of $1/2\pi$. In this particular case, the spin polarization of the excited state show oscillations with the frequency of the detuning $\omega_c-\omega_{hh}$ similar to those seen in the Kerr rotation.

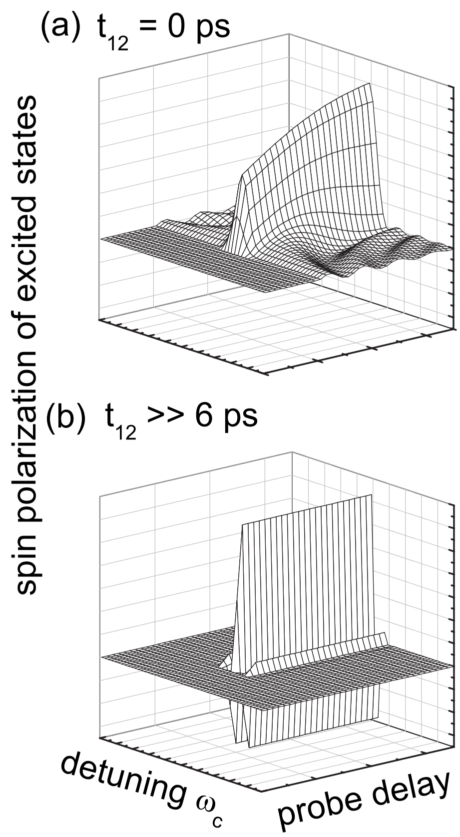

FIG. 8. Simulations of temporal transients for spin polarization of the upper state in a two-level system excited by polarization shaped laser pulses as a function of detuning $\omega_c-\omega_0$, where ω_0 is the frequency of the transition in the two-level system. All the relaxation processes including optical decoherence were neglected ($T_2\gg|t_{12}|$). (a) $t_{12}=0$ and (b) $t_{12}\gg 6$ ps.

Simulations also show that the phase of the beatings is directly related to the phase difference between the x- and y-polarized parts of the pump pulse, being in good agreement with the experiment. It should be noted that the absolute phase of the beatings extracted from the experiment is different from the one found theoretically. This is most likely explained in terms of many body effects, which are beyond the scope of this paper.

The agreement between theory and experiment clearly demonstrates that magneto-optical Kerr effect is strongly affected by off-resonantly excited spin populations of the electrons in the excited state. When circularly polarized off-resonant optical excitation is accompanied by an on-resonant linearly x-polarized pumping, quantum interference introduces beats in the laser-induced spin population of the excited state. The interference between off-resonantly and "resonantly" excited spin populations must be taken into account when the spin dynamics during the action of the laser pulse is analyzed. We could not find any indication for ultrafast spin relaxation due to intrinsic spin-flip mechanisms (for instance, hole-spin relaxation) nor for charge relaxation on the time scale of the laser pulses, except an exponential decay with a characteristic time of about 2 ps observed just above the exciton transition. The nature of this relaxation process will be an issue for future studies.

Finally, we would like to note that although we were able to generate circularly polarized fields having independent control of temporal and spectral resolution, the quantum beats do not allow us to overcome the Fourier limit in the optical control of spins.

However, if we consider spin system with even larger optical coherence times, an intriguing result is obtained in the simulations based on Eq. (2) when the x- and y-polarized parts of the pump do not overlap ($t_{12} \gg 6$ ps) and the optical decoherence time T_2 is large $T_2 \gg |t_{12}|$. In this case the quantum beats are suppressed [see Fig. 8(b)] and the spin polarization of the excited state exhibits a femtosecond rise time defined by the duration of the shortest pump pulse, while being observed only in the narrow spectral range defined by the longer pump pulse. Thus under these conditions, overcoming the Fourier limit in the optical control of spins may be feasible.[23]

IV. CONCLUSIONS

To conclude the photoinduced magneto-optical Kerr effect and spin dynamics that occur during the action of a picosecond polarized laser pulse is investigated in the spectral range exciton and trion transitions in a $CdTe/Cd_{0.78}Mg_{0.22}Te$ multiple quantum well. Using polarization pulse shaping technique and different shapes of the pump pulses we demonstrate that ultrafast evolution of the photoinduced Kerr effect during the excitation of a laser pulse cannot be explained by spin relaxation via an intrinsic spin-flip mechanism (hole-spin relaxation) alone. Instead, the spin population generated by off-resonant optical transitions is shown to have a considerable influence on the ultrafast dynamics of the magneto-optical Kerr effect. Although such an off-resonantly excited spin population is present only during the action of the laser pulse, quantum interference of "off-resonant" and on-resonant components leads to beats of the spin population of the excited state. The beatings are seen in the magneto-optical signal. The frequency and amplitude of the beatings are determined by the shape of the pump pulse.

ACKNOWLEDGMENTS

This work was partially supported by the Dutch National Initiative NanoNed (Nanospintronics), Nederlandse Organizatie voor Wetenschappelijk Onderzoek (NWO), EU programs INTAS, ITN FANTOMAS, NMP UltraMagnetron, the Russian programs (RFBR, Nanostructures, and Spintronics), the Polish Ministry of Science and Higher Education (Grant No. 202 054 32/1189), and the Foundation for Polish Science (Grant No. 12/2007).

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