

# Implementation of double-pulse laser control in optical Kerr effect spectroscopy

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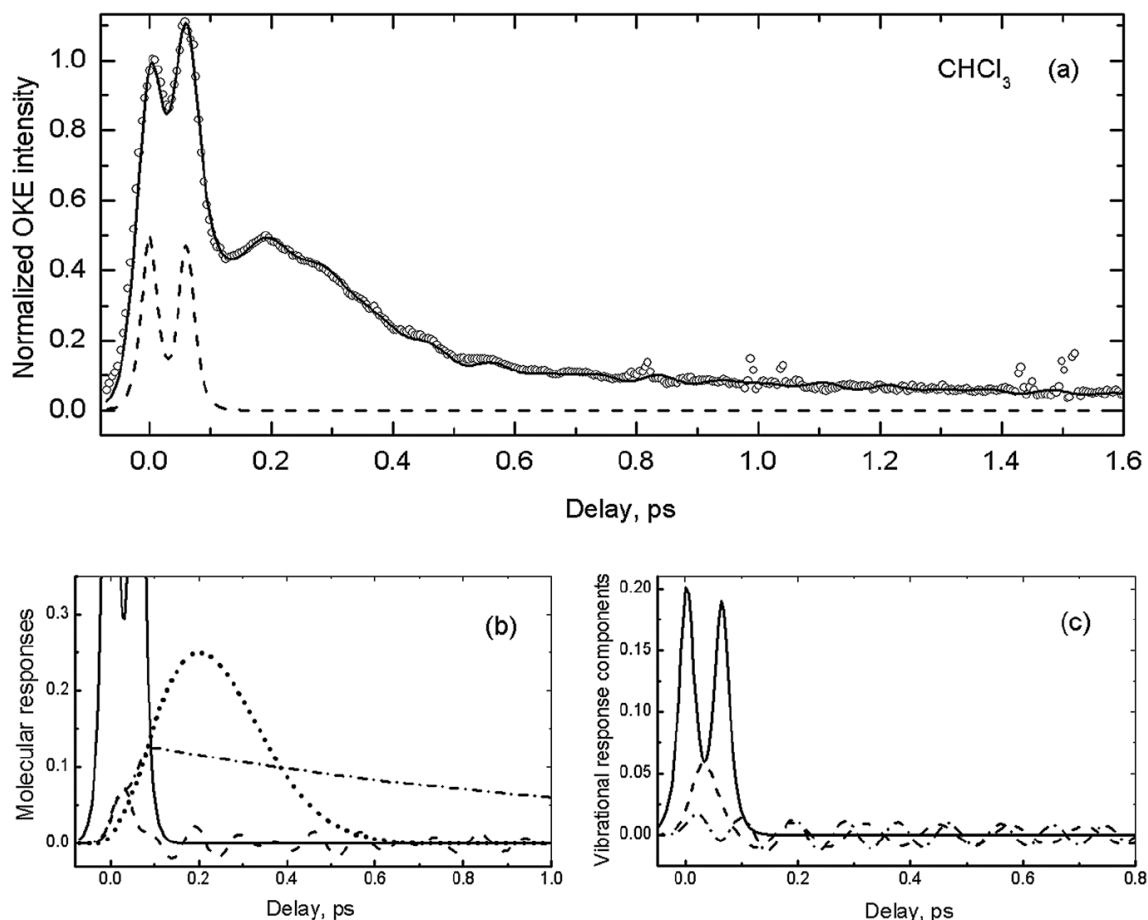
**Abstract.** Two-pulse control of time-dependent anisotropy in liquid  $CCl_4$  and  $CHCl_3$  at room temperature is implemented using femtosecond polarisation spectroscopy. Non-resonant excitation was enhanced by means of the double-pulse pump-probe technique. It is shown that by varying the delay between the exciting pulses and their relative intensity, selection of contributions of individual intramolecular modes into the recorded signal is achieved. The molecular responses were detected using the time-resolved optically heterodyne-detected optical-Kerr-effect technique.

## 1. Introduction

Laser control of the medium's state is one of the research priority areas focused on theoretical and experimental investigations [1]. Currently, the most effective instrument for the implementation of laser control is femtosecond technique, which makes it possible to excite a system by pulses comparable (in duration) to the period of molecular oscillations [2, 4, 3]. One of the laser control schemes is based on the use of multipulse excitation. The wave packets in the medium are formed by timed sequences of femtosecond pulses [5]. The key to excite a certain state of the medium is the optimized parameters of the multiple-pulse excitation (the pulse intensities, the delays between the pulses, etc.).

In this study we use the double-pulse non-resonant excitation to control the amplitudes of the individual modes in the vibrational responses of the molecules in liquid. We suppose that because of the short relaxation time of the coherent molecular motions (of less than or a few picoseconds), this technique was not employed in liquid at room temperature. In general, the pump pulse provides the vibrational and rotational wave packets due to which the anisotropy of the polarizability is induced in liquid. The evolution of the polarizability is probed by the time-resolved optically heterodyne-detected optical Kerr effect (OHD-OKE) [2, 3, 4]. The femtosecond pulses were generated by a standard self-mode-locked Ti:Sa laser (Avesta-Project, Troitsk, Russia). The average energy of the laser pulses on the carrying wavelength of 790 nm was  $4 \times 10^{-9}$  J. This pulses can't excite modes higher than  $500\text{ cm}^{-1}$ . In our experiment the two pump pulses are independently adjustable, and a standard scheme of the OHD-OKE transient detection [2, 3] was used: the probe beam polarization was at  $45^\circ$  with respect to the pump beam polarization, the intensity of the probe beam passed through the polarizer, the sample





**Figure 1.** The double-pulse excitation of the OHD-OKE signal. Delay between the pump pulses is 60 fs and their intensity ratios 0.94. (a)  $\circ$  – the experimental data, solid line – the simulation, dashed line – the envelope of the double-pulse intensities. (b) Solid line – the electronic response, dashed line – the vibrational response, dotted line – the librational response, dash-dotted line – the orientational response. (c) Solid line – the envelope of the double-pulse intensities, dashed line – the vibrational response component of the  $258\text{ cm}^{-1}$  mode, dash-dotted line – the vibrational response component of the  $362\text{ cm}^{-1}$  mode.

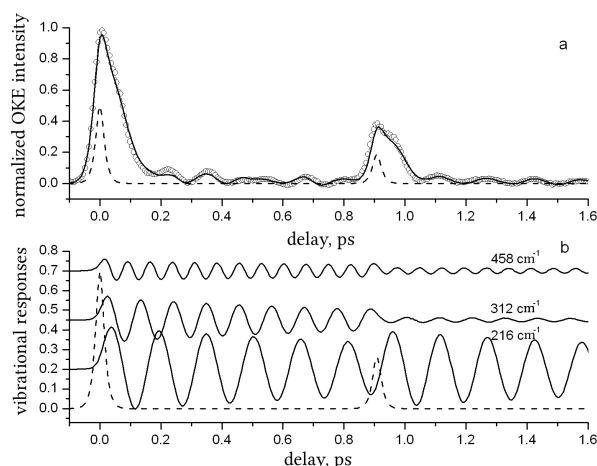
and the analyzer was measured. The optical heterodyne detection and the lock-in amplifier were used. We also used simulation of motion responses described in [5, 6].

## 2. Results

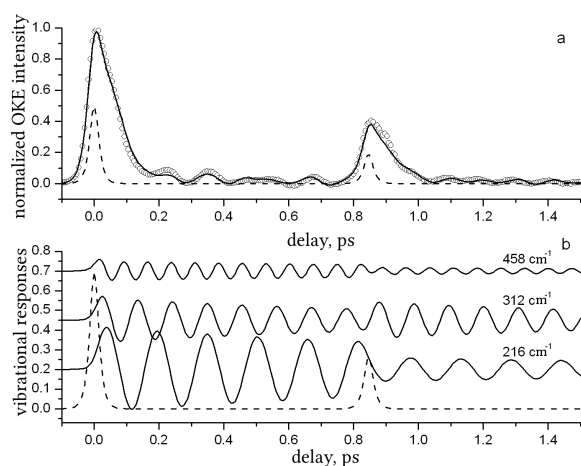
Chloroform has two Raman-active vibrational modes at frequencies less than  $400\text{ cm}^{-1}$ :  $258\text{ cm}^{-1}$  and  $362\text{ cm}^{-1}$ . One can see suppression of  $362\text{ cm}^{-1}$  mode by second pulse in figure 1. Registered signals are superposition of responses of individual motions [3]: instantaneous response of electrons and vibrational, librational and orientational response of nuclear skeleton of molecule.

By varying time delay between pump pulses and their relative intensity we can suppress or enhance responses of individual motions.  $\text{CCl}_4$  has three low-frequency Raman-active vibrational modes:  $458\text{ cm}^{-1}$ ,  $312\text{ cm}^{-1}$  and  $216\text{ cm}^{-1}$ . One can see suppression of responses of individual vibrational modes in figure 2, figure 3 and figure 4. In the figure 2 one can see that responses of individual vibrational modes after second pulse excitation changes in 1.8

( $216\text{ cm}^{-1}$ ), 0.2 ( $312\text{ cm}^{-1}$ ) and 0.5 ( $458\text{ cm}^{-1}$ ) times. With the same relative intensities of pump pulses, but with time delay 845 fs between pulses after second excitation responses changes in 0.2 ( $458\text{ cm}^{-1}$ ), 1.9 ( $312\text{ cm}^{-1}$ ) and 0.5 ( $216\text{ cm}^{-1}$ ) times (figure 3). With the time delay of 1025 fs responses changes in 0.6 ( $216\text{ cm}^{-1}$ ), 0.2 ( $312\text{ cm}^{-1}$ ) and 2 ( $458\text{ cm}^{-1}$ ) times (figure 4).



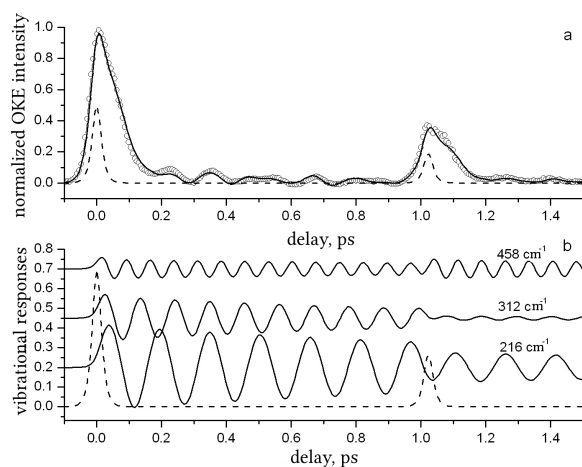
**Figure 2.** OKE signal in liquid  $CCl_4$  under double-pulse excitation, delay between pump pulses 908 fs, intensity ratio 0.38): (a)  $\circ$  – the experimental data, solid curve – simulation, dashed curve – envelope of the intensity of the exciting pulse. (b) simulation results of the vibrational responses, dashed curve is the envelope of the intensity of the exciting pulse.



**Figure 3.** OKE signal in liquid  $CCl_4$  under double-pulse excitation, delay between pump pulses 845 fs, intensity ratio 0.38): (a)  $\circ$  – the experimental data, solid curve – simulation, dashed curve – envelope of the intensity of the exciting pulse (b) simulation results of the vibrational responses dashed curve – envelope of the intensity of the exciting pulse.

### 3. Conclusion

These experiments present an example of the double-pulse controlling of the coherent molecular vibrational dynamics in liquid at room temperature. The laser control makes it possible to manipulate the vibrational responses in the OHD-OKE signal. The key parameters of the double-pulse scenarios increase or decrease the amplitude of the vibrational response and separate the desired modes in vibrational response are time delay between pump pulses and their relative intensity. Vibrational response of the mode  $458\text{ cm}^{-1}$  is not registered, because probe pulse width is  $\propto 400\text{ cm}^{-1}$ . However, simulation indicates that the double-pulse excitation also controls the intensity of the response of the given mode. Another important fact is suppression of the vibrational mode response at delays comparable to the relaxation time requires a decrease in the intensity of the second pump pulse. Note also that in liquids, where intensive rotational-vibrational responses are registered along with vibrational responses in the OKE signal, the scenario of complete suppression of the vibrational response at the lowest possible delay between



**Figure 4.** OKE signal in liquid  $CCl_4$  under double-pulse excitation, delay between pump pulses 1025 fs, intensity ratio 0.38): (a)  $\circ$  – the experimental data, solid curve – simulation, dashed curve – envelope of the intensity of the exciting pulse (b) simulation results of the vibrational responses dashed curve – envelope of the intensity of the exciting pulse.

the exciting pulses can be used to extract the rotational response. The contributions due to the molecular rotations increase as a result of the impact of the second pump pulse at any delay.

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