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Ultrafast Intermolecular Motions in Liquids Using the Optical Kerr Effect

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Abstract. In this work we explored ultrafast molecular dynamics of liquids by the example of organic solvents. We used two pump pulses with orthogonal linear polarization for the separate measurement of the interaction induced response in liquids through optically-heterodyne-detected optical-Kerr-effect (OHD-OKE). The pulse duration, the delay between pulses and the relation between the pump pulses amplitudes are the key parameters to manipulate inter- and intramolecular responses in OHD-OKE signal.

Keywords: multiple-pulse spectroscopy, coherent control, ultrafast optical Kerr effect, femtosecond spectroscopy, molecular dynamics in liquid.

Optically-heterodyne-detected optical Kerr effect (OHD-OKE) is a popular tool for monitoring the ultrafast dynamics of liquids [1]. Fast femtosecond technique progress makes it possible to detect the molecular responses within the frequency range of approximately 0-150 cm-1 by the typical femtosecond lasers. It is clear that other methods (for example, Raman scattering) yield the same spectra, but the OHD-OKE spectroscopy reveals a rather complete picture of the subpicosecond and picosecond orientational molecular motions, along with a less well characterized contribution to the relaxation from the intermolecular interactions.

The OHD-OKE optical experiments measure the third-order response function of the sample, which comprises two components: an effectively instantaneous electronic contribution and nuclear response functions [2]. The nuclear response arise from the intramolecular coherent vibrations, the molecular reorientations, the intermolecular translational, rotational collisions and the molecular librations - the fastest intermolecular motions, which occur as the molecule is torqued by the incident electric field [3].

To avoid the difficulty of spectra analysis we propose the using of high-order nonlinear optical technique permitting the separate measurements of different molecular processes [4]. For example, a multiple-pulse excitation was used to control the coherent phonon oscillations in a number of solid materials [5]. Recently the control of the coherent vibrational and rotational molecular motions in chloroform CHCl3 and acetonitrile C2H3N was achieved by the double-pulse excitation [6, 7]. In this paper we demonstrate the implementation of the double-pulse technique to separate intra- and intermolecular responses from the fifth-order OHD-OKE signal based on the coherent optical control principles. Pump pulses have 35 fs duration at 790 nm, less than 5×10-9 J. The pump pulses with orthogonal polarization excite the molecular motions non-resonantly, the medium is monitored by the

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third probe pulse, and the OHD-OKE response comprises the contributions of the low-frequency Raman-active vibrational modes, the orientational anisotropy, the librations and the electron hyperpolarizability of the molecules.

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