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Measurement of coherent polarisation relaxation times in condensed media using femtosecond interferometric spectroscopy

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Abstract. **An ultrafast spectrometer with a temporal resolution of 20 fs for studying the polarisation wave interference in condensed matter upon resonance excitation by femtosecond laser pulses is described. The transverse relaxation times $T_2 < 20$ and 50 fs are measured in optical media with considerably different inhomogeneous broadenings.**

Keywords: *femtosecond pulses, interferometric spectroscopy*.

The study of incoherent (excited-state population relaxation) and coherent (dephasing of the induced macroscopic polarisation) fast photoinduced processes in solids is performed using lasers generating ultrashort pulses with duration shorter than the characteristic times of processes involved. The dephasing of excited dipoles with frequencies within an inhomogeneously broadened line of a solid may occur at room temperature with characteristic times down to $10^{-13}-10^{-14}$ s. Various methods are used for studying fast processes, for example, the pump – probe technique, which is employed for measuring the dynamics of variation of the reflection or absorption in an excited sample [1]. However, experimental difficulties are encountered in the latter case due to the emergence of a spurious signal in the vicinity of zero delay [2], i.e., just in the case when the characteristic times of the processes being studied are comparable with the duration of the laser pulse, which is very important in direct measurements of the characteristic dephasing times T_2.

Such processes are studied using the interferometric techniques with a femtosecond temporal resolution, which provides information about the phase [3, 4]. The coherent processes can be detected easily and precisely with the help of the femtosecond linear optical correlation technique. This kind of an experiment is much simpler than the standard pump-probe technique and can be used to detect very weak signals or in case of a weak excitation power.

The method of polarisation wave interference in condensed media upon resonance excitation by femtosecond laser pulses consists in the following. A femtosecond pulse is split by a Michelson interferometer into two identical collinear pulses having the same intensity and polarisation. On leaving the interferometer, these two pulses fall on the sample by inducing in it two coherent polarisation waves, which interfere with each other if the delay time is comparable with T_2. The obtained interference pattern (depen-dence of the radiation intensity on the delay between the two pulses incident on the sample) is detected with a sensitive detector. The range of times T_2 being measured is detemined by the duration of the laser pulse used in the experiment. In the layout proposed by us, this duration does not exceed 20 fs, which allows us to measure time periods an order of magnitude shorter than those measured in [3, 4].

To realise the method described in this work, we used a femtosecond Ti:sapphire laser pumped by a 4-W cw Spectra-Physics argon laser having instability of below 1 %. The pulse repetition rate was determined by the round-trip time of a pulse in the laser cavity and was 82 MHz. The output pulse duration was about 60 fs for a spectral half-width $\sim 60-70$ nm with a maximum at 800 nm. An external two-prism compressor was used for pulse shortening below 20 fs for an average pulse power of about 100 mW.

After the compressor, the laser beam was split in the Michelson interferometer by a semitransparent mirror into two beams, one of which passing through a delay line (Fig. 1). The delay line consisted of a totally reflecting mirror mounted on the micrometer table for a rough equalisation of the interferometer arms, and of a precision delay device in the form of a 2-mm thick plane-parallel quartz plate mounted on a rotary table. The turning of the plate (continuos variation of the pulse delay) was performed by a step motor controlled by a computer, with an accuracy better than 0.15 fs.

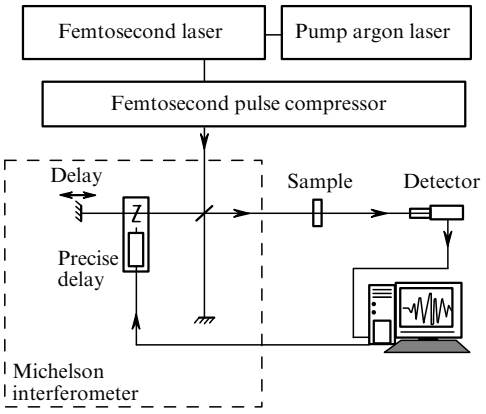

Figure 1. Schematic of an interference spectrometer.

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The samples under study were placed at the interferometer output directly in front of a synchronous detector measuring the dependence of the intensity of the interference pattern on the delay. As the test samples, we used standard optical filters PS-7 and SZS-21 having the absorption maximum at the laser wavelength (near 800 nm), but with strongly different widths of the inhomogeneous absorption bands (the half-width $\sim$40 nm of the PS-7 absorption band was much smaller than the half-width $\sim$500 nm of the SZS-21 absorption band).

It follows from the dependences (Fig. 2) that the time T_2 for the broadband filter SZS-21 does not exceed the laser pulse duration because the interference pattern remains virtually unchanged when a filter is placed at the interferometer output (cf. Figs 2a and 2b). The opposite situation is observed for the PS-7 filter (Fig. 2c). In this case, the time T_2 can be estimated by using the solution of Bloch optical equations for a two-level system at low excitation energies in the approximation of a slowly varying pulse envelope [4]. The detected signal as a function of the delay time t_d has the form

$$I(t_d) \sim 1 + \cos(\omega_0 t_d) \exp(-t_d/T_2), \quad (1)$$

where ω_0 is the resonance frequency, i.e., the observed interference pattern represents cyclic oscillations with frequency ω_0, which decay exponentially with the decay time T_2. Taking into account that $1/\omega_0 \approx 2.7$ fs in our case, T_2 for the PS-7 filter can be estimated as $\sim$50 fs from the decay of the interferogram envelope shown in Fig. 2c.

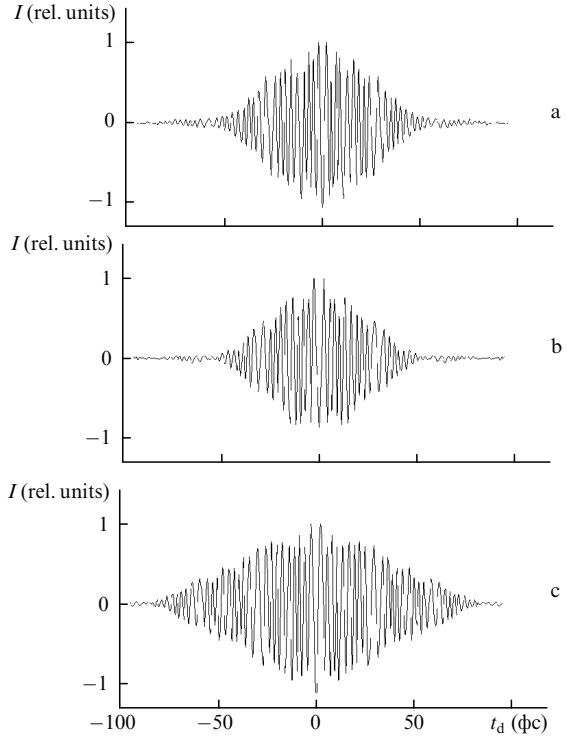

Figure 2. Experimental interferograms without sample (a), for SZS-21 (b), and for PS-7 filters (c).

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