Electro-Optic THz-TDS Based on Laser Pulse Spectrum Changes

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Abstract—**Novel schemes for THz time-domain spectroscopy (THz-TDS) are suggested and approbated. Substantial improvement of the sensitivity compared to the standard electro-optic (EO) detection technique (at high frequencies) and to the previously shown technique based on laser pulse energy changes is demonstrated in experiment.**

I. INTRODUCTION

CRYSTALS are widely used for THz-TDS based on Pockels effect.[1] Under this effect terahertz wave radiation (TR) can influence polarization of femtosecond laser pulses that is used for terahertz waveform measurements. However, TR in EO crystal can also influence energy,[2-4] spatial distribution[5] and spectrum of the laser pulses.[4] In the report, we present our recent theoretical and experimental study of new THz-TDS schemes based on the changes of the laser pulse spectrum.[6]

II. RESULTS

When a femtosecond optical pulse and a terahertz pulse co-propagate in a nonlinear EO crystal the nonlinear-optical interaction gives rise to changes both in the amplitude ΔA_ω and in the phase $\Delta\varphi_\omega$ of the output optical field.[2-4] Conventional EO sampling ellipsometry schemes are based on Pockels effect (determined by the phase modulation $\Delta\varphi_\omega$ of polarization components of the optical pulse).[1] The amplitude variation ΔA_ω leads to the change of the total laser pulse energy ("probe-energy" EO sampling)[2-4] and to the change of the pulse spectrum.[4] The "probe-energy" scheme can be useful in a number of cases considered before,[2-4] but for the same TR electric field and in the same EO crystal the magnitude of the registered signal is weak compared to the standard detection technique.[3,4]

However, the change of the total laser pulse energy is directly connected with the shift of the laser pulse spectrum.[4] This spectrum shift can be used for considerable improvement of the sensitivity of the "probe-energy" scheme by placing an edge filter between EO crystal and a photo-detector. The resulting response R of this scheme depends on the OR spectrum and on the transmission coefficient of the edge filter T_ω. For small amplitude changes ($\Delta A_\omega \ll A_\omega$), the response R can be described as

$$R \sim \int T_\omega (2A_\omega \Delta A_\omega + A_\omega^2)d\omega .$$

The value of ΔA_ω is proportional to $\omega(A_{\omega+\Omega} - A_{\omega-\Omega})$.[4] The value ΔR of the TR-induced change of the response, which is determined as a difference between the responses for the non-zero and zero ($R_0 \sim \int T_\omega A_\omega^2 d\omega$) amplitudes of the input TR electric field, is proportional to

$$\Delta R \sim \int T_\omega A_\omega \Delta A_\omega d\omega \sim \int T_\omega \omega A_\omega (A_{\omega+\Omega} - A_{\omega-\Omega})d\omega .$$

Here, the sign of the term $\omega A_\omega(A_{\omega+\Omega} - A_{\omega-\Omega})$ changes when the frequency varies along the laser pulse spectrum. If the frequency dependence of T_ω can be neglected ("probe-energy" EO sampling) the results of integration below the central frequency of the laser pulse spectrum ω_0 and above it partly compensate each other (it can be also interpreted in terms of the mutual compensation of the difference-frequency $(\omega-\Omega)$ and sum-frequency $(\omega+\Omega)$ generation processes[5]). That is the reason why the relative change of the signal $\Delta R/R$ ($\approx \Delta R/R_0$) is rather weak in the "probe-energy" EO sampling scheme.[4] When the edge filter is used, the optical energy passes through the filter primarily at frequencies higher (lower) than its cut-on (cut-off) frequency. For an edge filter with a cut-on (cut-off) frequency higher (lower) than ω_0, only the optical waves with frequencies within the intervals where the sign of the term $\omega A_\omega(A_{\omega+\Omega} - A_{\omega-\Omega})$ does not change are incident on the photo-detector. Thus, ΔR increases. Also, the value of R decreases, as the edge filter transmits only part of the initial pulse spectrum. Both these factors increase the $\Delta R/R$ ratio. The results of model calculations for different values of the cut-on (cut-off) frequency are presented in figure 1. It is seen that the response of the scheme with an edge filter grows when the cut-on frequency shifts from the central frequency of the laser pulse spectrum and can overcome the response of the standard EO scheme.
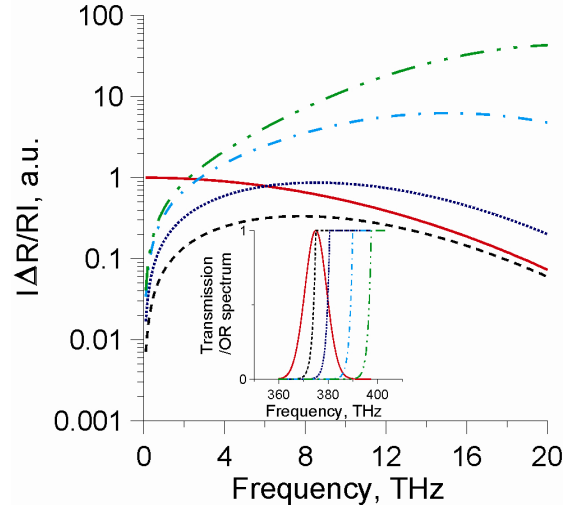

Fig. 1. The main graph represents responses |ΔR/R| of the standard measurement scheme (red solid line) and of the schemes with different edge filters (the other curves). Transmission curves of the edge filters according to the laser pulse spectrum are presented in the inset. The responses of the scheme with an edge filter in the main graph correspond to filters with transmission curves in the inset with the same line type.

In experiment the increase of the response more than in twenty times was observed comparing with the "probe-energy" scheme (figure 2). The signal from the studied scheme was also very close to the time derivative of the signal received in the standard scheme, in agreement with the results of Ref. 4. The increase of sensitivity was observed when the filter cut-on frequency was adjusted to a certain optimal value. In figure 2(b) it is seen that, even for the same EO crystal, the response of the studied scheme is comparable to that of the standard scheme at high TR frequencies (~7 THz). However, further increasing of the cut-on frequency led to reduction of the registered signal amplitude and to significant changes in the observed waveforms. We attribute these changes to unsatisfactory wave synchronism between the TR and the OR and to strong deviation of the initial laser pulse shape from the spectrally limited Gaussian pulse at the side band frequencies.
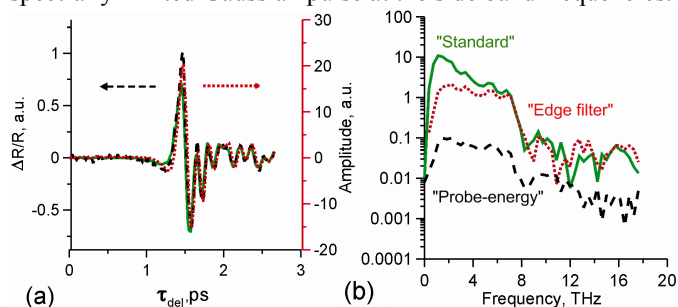
Fig. 2. (a) Terahertz induced signals $\Delta R/R$ for different time delays detected by three measurement units and 200μm GaP crystal. The green-solid/black-dashed/red-dotted lines are obtained using the [standard EO sampling]/"probe-energy" scheme]/[scheme with the edge filter]. The time dependence measured by unit (b) is processed by taking time derivative. Its vertical value does not match the scales of the axes. (b) Fourier amplitude spectra of unprocessed time dependences.

To achieve further improvement of the sensitivity the scheme was modified. The modification consisted in placing an edge filter into the probe beam path before the EO crystal and in measuring after the crystal the TR-induced changes of the optical wave energy at the frequencies corresponding to the slope of the filter. The change of the optical wave intensity $\Delta I_\omega/I_\omega$ at these frequencies may be very high for sharp frequency dependence of the transmission T_ω of the filter.[6]
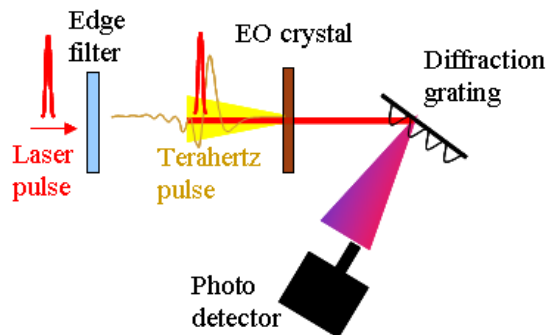
Fig. 3. Schematic of the experimental setup with the edge filter and diffraction grating.

To utilize these changes in experiment, the laser pulse transmitted through the edge filter and the EO crystal was decomposed in the frequency spectrum by a diffraction grating and the energy changes of the optical spectral components located at the slope of the filter were measured by a photodetector (figure 3). The detection sensitivity achieved in experiments with modified optical spectrum and diffraction grating was much better at high TR frequencies in comparison with the standard EO scheme (figure 4).
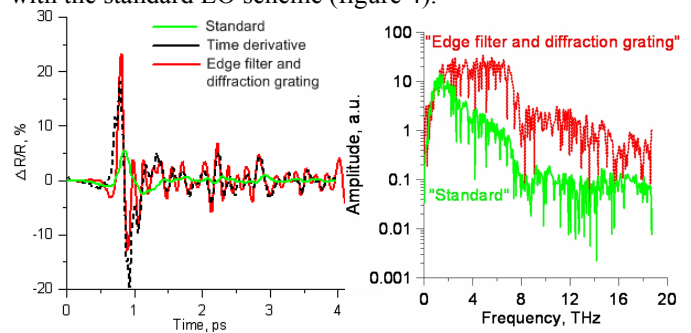
Fig. 4. Terahertz induced signals $\Delta R/R$ for different time delays and their Fourier amplitude spectra detected by two EO sampling techniques and 200μm GaP crystal. The green/red lines are obtained using the [standard EO sampling]/[sampling with the edge filter and diffraction grating]. Black line – the time dependence measured by the standard technique and processed by taking time derivative (its vertical value does not match the scale of the axis).

III. Summary

In conclusion, we combined the "probe-energy" scheme with spectral filtering utilizing laser pulse spectral changes that accompany TR-induced variations of its energy. According to our results, this method enables increasing the sensitivity of time-domain spectroscopy at high TR frequencies by more than an order of magnitude compared to the standard EO detection technique. In experiments, we used GaP and ZnTe[6] crystals that readily fit both standard and the studied schemes. The schemes with optical spectral filtering can also be used with other crystals with large natural birefringence and strong nonlinearity. For example, they can be used with a sandwich-structure[5] and periodically poled Mg:LiNb0$_3$ crystals,[2] which is promising for broad band and frequency-tunable narrow band TR detection with high sensitivity.

References

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