Dynamic grating mediated energy transfer between two sub-picosecond pulses in LiNbO$_3$

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Abstract. Two ultrashort pulses that interact in a nonlinear medium can excite a grating of the nonlinear absorption and refraction and their characteristics can be modified as a result of selfdiffraction from this grating. In this work, we adress an energy transfer between pulses of different energies and/or different frequencies within the visible spectral range in congruent, nominally undoped LiNbO$_3$ aiming to reveal new features that distinguish this process from the known transient and steady-state coupling of cw laser beams.

1. Introduction

In this presentation we describe a study of the direct energy exchange between ultrashort femtosecond pulses in LiNbO$_3$, the crystal which possesses multiple quite pronounced optical nonlinearities. Apart from the instantaneous high-frequency Kerr nonlinearity and two-photon absorption nonlinearity [1], several inertial nonlinear responses have been revealed including those which are due to photoexcitation of free carriers and formation of various kinds of polarons [2]. This crystal was the first in which a photorefractive nonlinearity and grating-assisted intensity redistribution of the recording waves has been discovered, see, e.g., [3]. We undertake this study to estimate how strong the coupling of subpicosecond pulses can be in LiNbO$_3$ and to establish the underlying wave mixing process(es) that is(are) responsible for energy redistribution between the interacting pulses.

2. Experimental Setup

The femtosecond laser system consists of a Ti:Sapphire oscillator with amplifier as a pump source for two independent tunable Optical Parametric Amplifiers (OPAs) [cf. [1]]. For the study of frequency degenerate interaction, the output of a single OPA was divided by a beamsplitter, while the use of two OPAs allowed for introducing any desirable frequency detuning between the recording pulses.

In the experiment, the two light pulses impinge upon the sample in a plane normal to the axis of spontaneous polarization and are polarized along this axis to avoid possible contribution of space-charge (photorefractive) gratings in investigated coupling processes. The angle of the beam intersection in air was chosen to be less than $5^\circ$ to ensure a good spatial overlap of two 100 fs-long pulses. With the help of an optical delay line in one of two intersecting light beams the recording pulses could be perfectly well adjusted in time or deliberately mismatched with a controllable time delay. A double grating setup was used as a compressor/stretcher to change the chirp. This pulse chirp opens the possibility to introduce a frequency difference by changing the temporal mismatch, even in the case of the recording pulses with identical spectra.
3. Results & Discussion

Figure 1a shows the relative sample transmission \( T \) for different pulse delays \( \Delta t \) for two interacting pulses with identical pulse durations, pulse energies, chirp coefficients and spectra (centred at 590 nm). The upper and lower graphs differ in the sign of chirp (and pulse energy). In both graphs, pronounced energy redistribution (± 20 %) is observed at \( \Delta t \) close to FWHM of the pulses themselves. Within the time interval where pulses interact a red-shifted portion of one pulse is always amplified in expense of the blue-shifted portion of the other pulse. Thus, for a fixed \( \Delta t \) the direction of energy flow is inverted if the sign of the chirp becomes opposite. This type of pulse energy coupling via diffraction from a moving grating is already well known (see, e.g., for plasma gratings in gases [5]).

To confirm that it is just a frequency difference that governs this particular energy coupling process we used two pulses with 5 nm difference in wavelength. Instead of two, well separated in \( \Delta t \) energy coupling areas (Figure 1a) we obtained only one high coupling area centered at \( \Delta t = 0 \).

Figure 1. (a) Pulse delay dependence of transmission \( T(\Delta t) \) for two identical interacting pulses (see text) with pulse durations \( \tau = 200 \) fs (FWHM) and opposite chirp parameters \( C \). (b) Wavelength detuning \( \delta = \lambda_{\text{probe}} - \lambda_{\text{pump}} \) dependence of the gain \( G \) for a weak probe pulse.

The next set of experiments consisted in gain measurement of a low signal probe pulse in presence of a strong pump pulse with tunable frequency. Figure 1b shows that nonzero coupling gain of a weak probe exists also in case of strict degeneracy, when both, the pump and probe pulse have identical spectra centered at 488 nm. This confirms the presence of another coupling process that is sensitive not to the detuning in pulse frequencies but to pulse difference in intensity. We believe this is a manifestation of a transient energy transfer (TET) [5] known for coupling of long pulses in media with inertial nonlinearities but never considered before for femtosecond pulses. Thus the detuning dependence of Figure 1b consists of two contributions, a \( \delta \)-symmetric (odd function) dependence from TET and \( \delta \)-anti-symmetric (even function) contribution from coupling of frequency nondegenerate pulses.

The physical processes that are causing these optical nonlinearities are not yet established, but we know that for observing TET the decay time of the nonlinearity should be longer comparing to the pulse duration. However, for efficient coupling of frequency nondegenerate pulses, on the opposite, this time should be comparable or shorter than the pulse duration.

Acknowledgements The financial support of the Deutsche Forschungs Gemeinschaft (DFG) via projects IM37/5-2, INST 190/137-1 FUGG and INST 190/165-1 FUGG is gratefully acknowledged.

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