Temporal Analysis of a Mode-Locked Nd-Glass Laser by Four-Wave Mixing

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Abstract. The duration, shape, and background intensity level of pulses of a passively mode-locked Nd-glass laser are determined by noncollinear phase-matched four-photon frequency mixing of time-separated fundamental and second-harmonic pulses in a KI crystal. The pulse shape is found to be slightly asymmetric with slower trailing than rising parts. The background intensity level before and behind the main pulse is about 5×10^{-6} the peak pulse intensity.

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The application of mode-locked lasers in time-resolved spectroscopic studies requires an accurate analysis of the temporal behaviour of these lasers. Various techniques have been invented to determine the duration, shape, and background energy level of picosecond lasers. Frequently used methods for the measurement of pulse durations are the two-photon fluorescence, the second harmonic, and the Duguay shutter correlation techniques. The temporal shape and duration are measured with streak cameras. These techniques were reviewed in [1–3]. Since the publication of these reviews further techniques have been developed for the measurement of pulse durations [4–7], pulse shapes [8] and background energy contents [9, 10]. The shot-to-shot pulse fluctuations were analysed in [11].

In this paper we analyse the temporal behaviour of a mode-locked Nd-glass laser by noncollinear phase-matched, nonresonant four-photon frequency mixing of its fundamental and second-harmonic pulses. This technique was first applied by Auston [12]. Here the analysis is refined by application of four-photon frequency mixing theory to the determination of pulse duration, shape, and background intensity content. After calibration of the system the duration of picosecond pulses may be measured in single shots.

1. Method

The fundamental (frequency v_L) and second-harmonic (frequency v_P) wave of a mode-locked Nd-glass laser

interact in a nonlinear optical medium by four-photon frequency mixing $v_P + v_P - v_L \rightarrow v_S$. The energy conservation requires $v_S = 2v_P - v_L$. The angle φ between the interacting laser beams is adjusted to the phase-matching condition $\mathbf{k}_P + \mathbf{k}_P - \mathbf{k}_L = \mathbf{k}_S$. The pump pulses are time separated by an optical delay line and the energy conversion $\eta = W_S/W_L$ of laser light at frequency v_L to frequency v_S is measured versus delay time t_D ($t_D = 0$ at optimum temporal overlap of pump pulses in sample, $t_D > 0$ when pulse at v_L behind pulse at v_P). The schematic arrangement of the interacting light beams is depicted in Fig. 1.
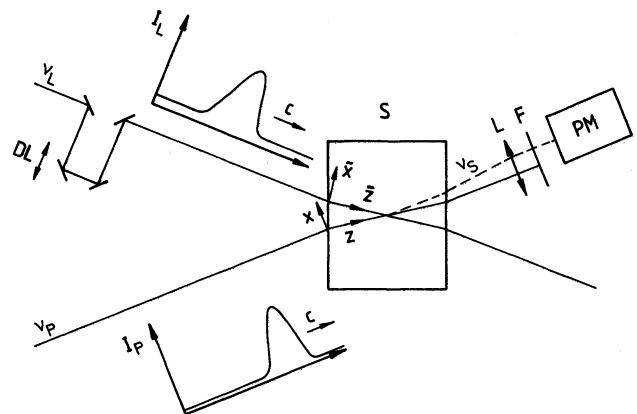

Fig. 1. Schematic arrangement of noncollinear phase-matched four-wave mixing. (DL: optical delay line; S: interaction medium; L: lens; F: filter; PM: photomultiplier tube)

The energy conversion η is a function of the peak pulse intensity I_{OP} (frequency ν_P), of the temporal and spatial intensity distributions, the delay time t_D, the phase-matching angle φ, the refractive indices n_L, n_P, n_S, the absorption coefficients α_L, α_P, α_S and the beam divergencies Θ_L, Θ_P [13]. The ratio of energy conversion $r(t_D) = \eta(t_D)/\eta(0)$ at a fixed pump pulse peak intensity I_{OP} is given by

$$r(t_D) = \frac{F(t_D)}{F(0)} \tag{1}$$

with [13]

$$F(t_D) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left\{ \int_0^l h_P\left(\frac{x}{x_P}\right) f_P\left(\frac{t'_P}{t_P}\right) \right.$$
$$\cdot \left[h_L\left(\frac{\tilde{x}}{x_L}\right) f_L\left(\frac{t'_L}{t_L}\right) \right]^{1/2} \exp\left[-\alpha_P z - \frac{1}{2}\alpha_L \tilde{z} \right.$$
$$\left. \left. - \frac{n_{gS}\alpha_S}{2n_S\cos(\psi)}(\tilde{l} - z) \right] dz \right\}^2 dx' dt' . \tag{2}$$

The small effect of the beam divergencies Θ_L and Θ_P on $r(t_D)$ is neglected. The parameters in (2) are [13]:

$x = x' - z\tan\psi$, $t = t' + n_{gS}^2 z/(n_S c \cos\psi)$,

$\tilde{x} = x\cos\varphi - z\sin\varphi + 2b\sin(\varphi/2)$,

$t'_P = t - n_{gP}z/c$, $t'_L = t - t_D - t_0 - n_{gL}\tilde{z}/c$,

t_0 is chosen that $F(t_D = 0)$ is maximal,

$\tilde{z} = z\cos\varphi + x\sin\varphi - 2b\tan(\varphi/2)\sin(\varphi/2)$,

$b = l/2$ (pump pulses overlap in center of sample),

$\tilde{l} = l/\cos(\varphi/2)$,

$\varphi = \arccos\{[(2n_P\nu_P)^2 + (n_L\nu_L)^2$
$\qquad - (n_S\nu_S)^2]/(4n_P n_L \nu_P \nu_L)\}$,

$\psi = \arcsin\{n_L\nu_L\sin\varphi/[(n_L\nu_L)^2$
$\qquad + (2n_P\nu_P)^2 - 4n_L n_P \nu_L \nu_P \cos\varphi]^{1/2}\}$.

l is the sample length; n_{gL}, n_{gP}, n_{gS} are the group refractive indices; c is the vacuum light velocity.
The temporal analysis of laser pulses at frequency ν_L is achieved by comparing calculated energy conversion ratios r with measured values. The spatial pulse shape is assumed to be Gaussian, i.e. $h_L(\tilde{x}/x_L) = \exp(-\tilde{x}^2/x_L^2)$, $h_P(x/x_P) = \exp(-x^2/x_P^2)$. The temporal pulse shape is choosen in the form of ($i = L, P$):

$$f_i(t'_i/t_i) = \exp(-t'^2_i/t^2_i)[1 - \Theta(t'_i)]$$
$$+ \{\cosh[t'_i/(\kappa_i t_i)]\}^{-2}\Theta(t'_i)$$
$$+ \delta_i[\Theta(t'_i + T/2) - \Theta(t'_i - T/2)] . \tag{3}$$

$\Theta(t'_i)$ is a step function [$\Theta(t'_i) = 0$ for $t'_i < 0$, $\Theta(t'_i) = 1$ for $t'_i \geqq 0$]. κ_i is a fitting parameter for the pulse asymmetry and δ_i represents the background intensity level. T is

the round-trip time of pulses in the laser resonator ($T = 10\,\text{ns}$). κ_L and κ_P are set equal to κ in the calculations.
In case of collinear four-photon frequency mixing of pump pulses at frequencies ν_P and ν_L in dispersionless and nonabsorbing media the energy conversion ratio $r(t_D)$ would simplify to

$$r(t_D, \varphi = 0) = G(t_D)$$
$$= \frac{\displaystyle\int_{-\infty}^{\infty} f_P^2(t/t_P) f_L[(t - t_D)/t_L]\, dt}{\displaystyle\int_{-\infty}^{\infty} f_P^2(t/t_P) f_L(t/t_L)\, dt} . \tag{4}$$

This correlation function was used in [12] for the pulse analysis. The deviation of $r(t_D, \varphi \neq 0)$ from $G(t_D)$ increases with phase-matching angle φ.

2. Experimental

The laser system consists of a passively mode-locked Nd-phosphate glass laser (laser glass Hoya LHG5, bleachable dye Eastman 9860). A single pulse is selected from the pulse train by a laser triggered Kerr shutter (opening time: 10 ns) and increased in energy by a laser amplifier. The second-harmonic pulse is generated in a KDP crystal of 1 cm length. The fundamental and second-harmonic pulses are separated with a harmonic beam splitter and directed to the nonlinear medium under the phase-matching angle. In the path of the fundamental laser beam an optical delay line is inserted. A KI crystal of 2 cm length is used as four-photon mixing sample (phase-matching angle in sample $\varphi = 25.42°$). Other nonlinear optical media might be used as well (e.g. water with $\varphi = 10.68°$). The nonlinear susceptibility of the medium has to respond instantaneously to the interacting beams. The generated light at frequency ν_S is detected with a photomultiplier tube. The geometrical arrangement of the light beams is depicted in Fig. 1. (For more details see [13, 14].)

3. Results

The energy conversion η is measured versus delay time t_D in the range between $-300\,\text{ps}$ and 1 ns. The energy conversions at different pump pulse peak intensities I_{OP} around $2 \times 10^9\,\text{W/cm}^2$ are normalized to a fixed peak intensity $\bar{I}_{OP}$ by using the proportionality $\eta \propto I_{OP}^2$, i.e. $\eta(\bar{I}_{OP}) = \eta(I_{OP})\bar{I}_{OP}^2/I_{OP}^2$. The measured conversion ratios $r(t_D)$ are shown as data points in Fig. 2. The solid curves are calculated by use of (1)–(3). The KI parameters are [13]: $n_L = 1.6358$, $n_P = 1.6778$, $n_S = 1.7625$, $n_{gL} = 1.6624$, $n_{gP} = 1.8067$, $n_{gS} = 2.2345$, α_L
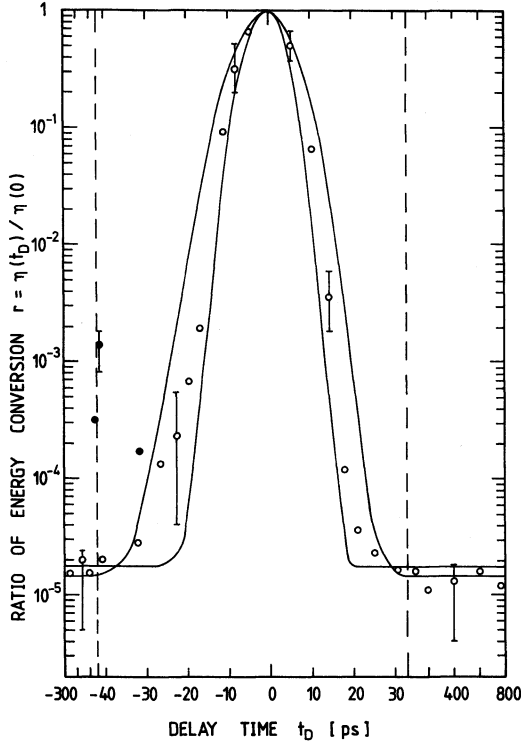
Fig. 2. Ratio of energy conversion versus delay time in KI ($l = 2$ cm). Open circles, measured average energy conversions, closed circles indicate influence of reflection of laser at frequency ν_L from an optical surface. Curves are calculated for rising Gaussian and decaying hyperbolic secant pulses (3) with $\kappa = 1.6$, $\delta_L = 5 \times 10^{-6}$, $\delta_P = \delta_L^2$, $\Delta t_L/\Delta t_P = 1.2$, $\Delta t_L = 3$ ps (inner curve, FWHM) and $\Delta t_L = 6$ ps (outer curve). Note change of time scale at dashed vertical lines

Fig. 3. (a) Calculated half-width of energy conversion curve versus pulse duration. Curve 1: nonlinear medium is KI; Curve 2: sample is H_2O; dashed curve is responsible for collinear interaction, (4). (b) Dependence of energy conversion on pulse duration. Interaction in KI (Curve a), in H_2O (Curve b). Dashed curve is valid for collinear phase-matched interaction. Pulse parameters as in Fig. 2 (curves nearly independent of asymmetry parameters κ)

$= 0.05$ cm^{-1}, $\alpha_P = 0$, $\alpha_S = 0.02$ cm^{-1}. The laser parameters are $\nu_L = 9480$ cm^{-1}, $\nu_P = 18,960$ cm^{-1}, $\nu_S = 28,440$ cm^{-1}, $x_L = 4.2$ mm, $x_P = 2.85$ mm and $\Delta t_L/\Delta t_P = 1.2$. The pulse durations (FWHM) are $\Delta t_L = 3$ ps (inner curve) and 6 ps (outer curve). The shape fitting parameter is $\kappa = 1.6$, the background to peak pulse intensity ratio is $\delta_L = 5 \times 10^{-6}$ and $\delta_P = \delta_L^2$.

Second-harmonic generation at small conversion efficiency in dispersionless media would produce pulses of duration $\Delta t_P = 2^{-1/2} \Delta t_L$ [15–18]. For our situation the ratio $\Delta t_L/\Delta t_P = 1.2$ was determined earlier [8].

The experimental points (open circles) fit well calculations for $\Delta t_L = 5$ ps, $\kappa = 1.6$ and $\delta_L = 5 \times 10^{-6}$. Each point represents an arithmetic average over 16 shots. The shot-to-shot fluctuations are indicated at some data points by bars which span the region from the forth lowest to the forth highest shot. It should be noted that reflections of the fundamental laser beam (frequency ν_L) at surfaces clearly show up in the post-pulse region ($t_D < 0$) when no antireflection coated or wedged optical components in the path are used. The full circles in Fig. 2 resulted from reflections at an uncoated cylindrical lens.
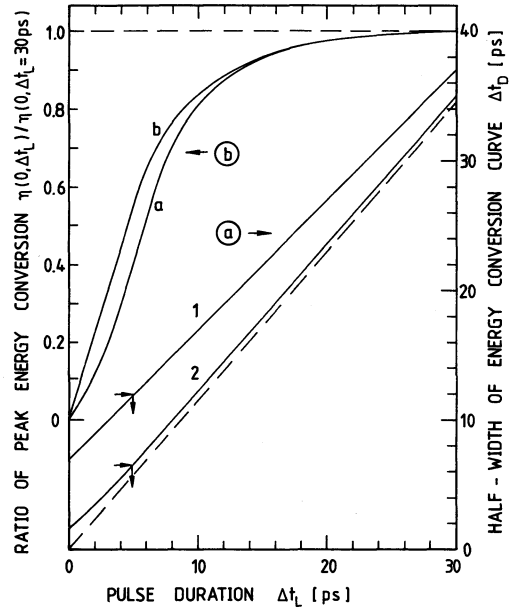
The temporal half-width Δt_D of $r(t_D)$ versus pulse duration Δt_L is shown in Fig. 3a. Curve 1 indicates the situation for KI where Δt_D is about 7 ps greater than Δt_L in the depicted region. This broadening results from the noncollinear propagation of the spatially extended (x_L, x_P) pump pulses which causes an elongation of the temporal pulse overlap. The experimental half-width of $\Delta t_D \simeq 12$ ps (Fig. 2) gives a pulse duration of $\Delta t_L \simeq 5$ ps. The correlation technique [$G(t_D)$ of (4)] leads to $\Delta t_D = 1.15 \Delta t_L$ (dashed curve). Curve 2 is responsible for the situation of noncollinear four wave mixing in water ($l = 2$ cm, $\varphi = 10.68°$, $n_L = 1.3247$, $n_P = 1.33468$, $n_S = 1.34815$, $n_{gL} = 1.33518$, $n_{gP} = 1.35783$, $n_{gS} = 1.39823$, $\alpha_L = 0.172$ cm^{-1}, $\alpha_P = 3.2 \times 10^{-4}$ cm^{-1}, $\alpha_S = 0.0023$ cm^{-1} [13]). In earlier investigations on four-wave mixing in water [14] a half-width of $\Delta t_D \simeq 6.5$ ps was found giving a pulse duration of $\Delta t_L \simeq 5$ ps.

The background intensity level δ_L is only approximately equal to the energy conversion ratio $r(|t_D| > 6\Delta t)$. The relation between δ_L and $r(|t_D| > 6\Delta t_L)$ depends on the pulse duration Δt_L as shown in Fig. 4 (KI, Curve 1; H_2O, Curve 2). The overlap of the pump pulses is reduced with decreasing pulse duration (group dispersion separates pulses). This fact causes a reduced energy conversion of the pulses (Fig. 3b, Curve a for KI, Curve b for H_2O) while the back-
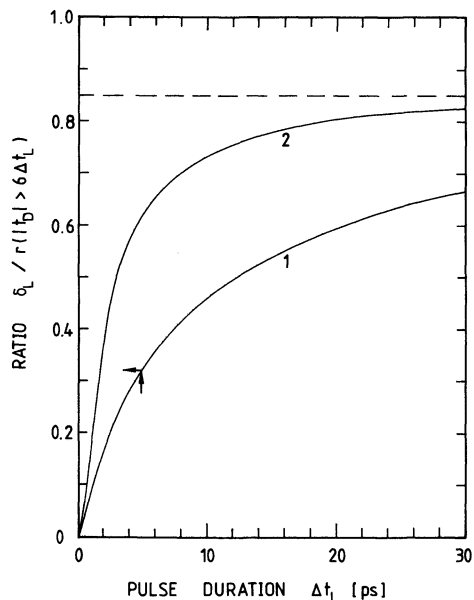
Fig. 4. Relation between background intensity level and energy conversion of background signal. Curve *1*: interaction in KI; Curve *2*: interaction in H_2O; dashed curve, result of collinear interaction, (4). Pulse parameters as in Fig. 2
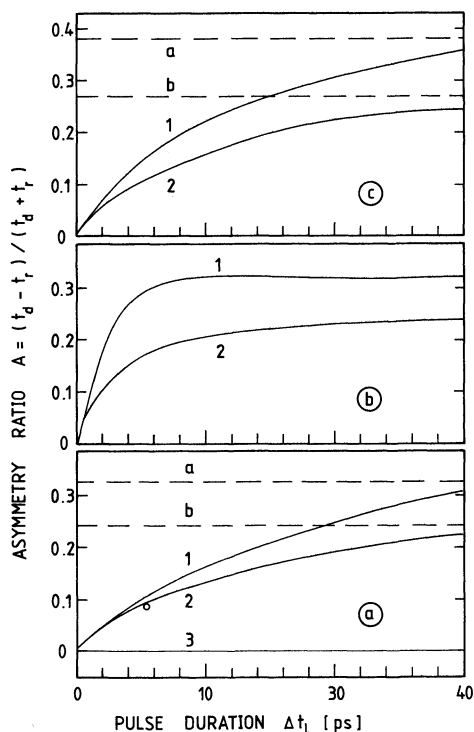

Fig. 5a–c. Asymmetry ratio of energy conversion curves versus pulse duration. (a) Interaction in KI; pulse duration ratio $\Delta t_L/\Delta t_P = 1.2$; (b) H_2O; $\Delta t_L/\Delta t_P = 1.2$; (c) medium KI, $\Delta t_L/\Delta t_P = 2^{1/2}$. Solid curves: *1*, $\kappa = 4$ (pulse asymmetry $A_L = 0.73$, similar defined as A); *2*, $\kappa = 1.6$ ($A_L = 0.43$); *3*, $\kappa = 0.6338$ ($A_L = 0$). Dashed curves represent result of correlation functions (independent of substance). a, $\kappa = 4$; b, $\kappa = 1.6$

ground signal at frequency ν_L interacts completely with the pump pulse at frequency ν_P. In our experiments it is $r(|t_D| > 6\Delta t_L) \simeq 1.5 \times 10^{-5}$ and the background intensity level is $\delta_L \simeq 5 \times 10^{-6}$. The background level is rather constant over the investigated time region (Fig. 2). The correlation function analysis [(4) dashed curve] gives a ratio of $\delta_L/r(|t_D| > 6\Delta t) = 0.85$ independent of the pulse duration Δt_L.

The asymmetry of the laser pulses causes an asymmetry of the energy conversion versus delay time. The asymmetry of $r(t_D)$ is lowered by the finite width Δt_P of the pump pulse at frequency ν_P and the elongated pulse overlap by the noncollinear arrangement. The asymmetry ratio $A = (t_d - t_r)/(t_d + t_r)$ is plotted versus pulse duration Δt_L in Fig. 5. t_d and t_r are defined as decay time and rise time by $\eta(t_D = -t_d < 0) = \eta(t_D = t_r > 0) = \eta(t_D = 0)/1000$. In Fig. 5a the curves belong to four-wave mixing in KI (*1*, $\kappa = 4$; *2*, $\kappa = 1.6$; *3*, $\kappa = 0.6338$) with a pulse duration ratio of $\Delta t_L/\Delta t_P = 1.2$. The experimentally observed asymmetry $A \simeq 0.09$ indicates a pulse asymmetry parameter of $\kappa \simeq 1.6$. In case of idealized forward four-photon interaction, (4), the asymmetry ratio is only reduced by the finite width of Δt_P (dashed curves; a, $\kappa = 4$; b, $\kappa = 1.6$). The curves in Fig. 5c indicate the influence of the pulse duration ratio $\Delta t_L/\Delta t_P$ (*1* and a: $\kappa = 4$; *2* and b: $\kappa = 1.6$; $\Delta t_L/\Delta t_P = 2^{1/2}$, medium KI). Figure 5b shows the asymmetry for four-wave mixing in water (*1*, $\kappa = 4$; *2*, $\kappa = 1.6$; $\Delta t_L/\Delta t_P = 1.2$).

After the optimum overlap position $t_D = 0$ and the energy conversion $\eta(t_D = 0)$ have been determined, the measurement of the energy conversion at a fixed delay time $\bar{t}_D$ allows the measurement of pulse durations in single shots. The pulse shape has to be assumed for these single shot measurements as it is necessary in the conventional two-photon fluorescence or second-harmonic correlation techniques.

The background energy content of the mode-locked laser may be reduced by passing the laser output through a saturable absorber. By putting a bleachable dye of small signal transmission $T_0 = 0.005$ (Eastman 9860) behind the laser amplifier, the background level decreased by a factor of 50 to $\delta_L \simeq 10^{-7}$.

The lowest measurable background intensity level is approached when the light generation at frequency ν_S by one of the pump pulses at either ν_L or ν_P alone becomes comparable to that by four-wave mixing of background intensity at ν_L with pump laser at ν_P. The fundamental laser at ν_L generates light at ν_S in forward direction by phase-mismatched third-harmonic generation. Curve *3* in Fig. 6 shows the energy conversion efficiency of third harmonic generation in forward
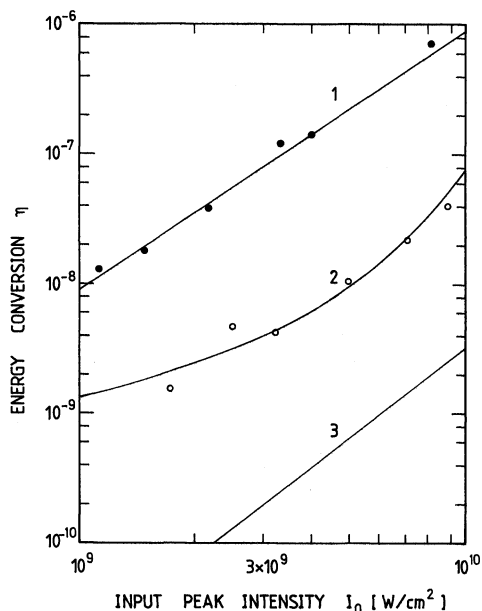
Fig. 6. Energy conversion by nonlinear optical processes in KI sample ($l=2$ cm). Curve *1*: four-photon frequency mixing $\nu_P + \nu_P - \nu_L \rightarrow \nu_S$ of background pulses at ν_L with pump pulse at ν_P. Closed circles, experimental points. Curve *2*: parametric four-photon interaction $\nu_P + \nu_P \rightarrow \nu_L + \nu_S$ of pump pulse at frequency ν_P. Open circles, experimental data. Curve *3*: phase-mismatched third-harmonic generation of pump pulse at frequency ν_L in forward direction

direction for KI as nonlinear medium ($l=2$ cm, $\chi^{(3)}_{xxxx}(-\nu_S; \nu_L, \nu_L, \nu_L)=4.8 \times 10^{-22}$ m^2/V^2 $=4 \times 10^{-14}$ esu [13], $\Delta k = 2.3 \times 10^4$ cm^{-1}, using [Ref 19, Eq. (22)]). The generated third-harmonic light does not enter the photomultiplier (Fig. 1) and is negligibly small compared to the signal due to background pulse interaction at ν_L with pump pulse at frequency ν_P (Curve *1*, full circles). The second-harmonic pump pulse generates light at frequency ν_S by phase-matched noncollinear parametric four-photon interaction $\nu_P + \nu_P \rightarrow \nu_L + \nu_S$ [19] (same phase-matching angle φ). The open circles indicate the measured energy conversion values $\eta = W_S/W_L = \alpha W_S/W_P (\alpha = W_P/W_L \simeq 0.2)$. The Curve *2* is calculated by use of [Ref. 19, Eqs. (11) and (13)] with $\chi^{(3)}_{xxxx}(-\nu_S; \nu_P, \nu_P, -\nu_L)=3\chi^{(3)}_{xxxx}(-\nu_S, \nu_L, \nu_L, \nu_L)=1.2 \times 10^{-13}$ esu [20], effective sample length $l_{eff} = 1$ cm, width of the generated light $\Delta\nu=40$ cm^{-1} (half-width of interference filter) and solid angle $\Delta\Omega = 5 \times 10^{-4}$ sr (adjusted to experimental points). In the intensity region around $I_{OP} \simeq 2 \times 10^9$ W/cm^2 the

parametric four-photon light is about a factor of 20 below the background signal level of fourphoton frequency mixing.

Conclusions

Noncollinear phase-matched four-photon frequency mixing was used to analyse the temporal behaviour of a mode-locked Nd-glass laser. The technique allows to measure the duration, shape, and background content of picosecond lasers with high accuracy. Pulse duration measurements in single shots are possible. Any transparent liquid or solid with reasonable electronic nonlinear susceptibility may be used as interaction medium. Media with small phase-matching angles (e.g. water) are preferable for the analysis of pulses with short duration ($\Delta t_L < 3$ ps).

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