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Measurement and compensation of frequency chirping in pulsed dye laser amplifiers

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Abstract. Rapid changes of the refractive index in the active medium of a pulsed, excimer laser pumped dye laser amplifier were investigated with an optical heterodyne technique. Time-dependent shifts in the phase of optical light waves could be observed which for Coumarin 102, 153 and 307 dyes at wavelengths of 488 and 550 nm depend predominantly on the thermal properties, in particular, on the expansitivity of the dye solutions. A fast Electro-Optic Modulator (EOM) using $LiTaO_3$ crystals has been designed for compensating these phase perturbations. With appropriately shaped voltage pulses applied to the EOM the resulting changes in the instantaneous frequency of the pulsed laser output could be lowered by a factor of 4 with respect to the values measured in the uncompensated light.

The amplification of cw laser light in a pulsed dye laser amplifier is a commonly used technique to produce pulsed radiation with high peak intensities and a nearly Fourier limited spectral distribution for a variety of spectroscopic applications. This method is well suited for nonlinear processes like, e.g., for inducing two-photon transitions or for frequency doubling of laser light.

In fact, the first approaches to measure the $1S$–$2S$ transition frequency in atomic hydrogen by Doppler-free two-photon spectroscopy relied on the availability of pulsed radiation [1, 2]. Higher accuracies could later on be achieved with laser light sources based on pulsed amplification of cw laser beams [3, 4]. The output of a single mode dye laser at a wavelength of 486 nm was amplified in a pulsed dye amplifier and then doubled in frequency in a nonlinear crystal in order to obtain radiation at $\lambda = 243$ nm for exciting the metastable $2S$ level from the $1S$ ground state. The absolute transition frequency could be determined at a relative accuracy of 10^{-8} [4] and a precise value for the Rydberg constant was extracted from the measurement. The $1S$ Lamb shift was obtained from a comparison of the $1S$–$2S$ transition frequency with the $2S$–$4P$ frequency difference. Both measurements comprised important contributions to metrology and to tests of bound-state Quantum ElectroDynamics (QED). The resolution achieved in these experiments was limited by phase fluctuations of the pulsed amplified light caused by rapid changes of the refractive index in the dye solution within the duration of the pulse [5]. The average frequency of the pulsed laser light was systematically blue-shifted with respect to the cw seed radiation by some 10 MHz; the effect is known as frequency chirping. Corrections needed to be applied based on the assumption of an average shift, which was usually measured by interferometry [6].

Another important example for pulsed amplification of cw laser light is the measurement of the 1^3S_1–2^3S_1 interval in positronium (e^+e^-) by Doppler-free two-photon spectroscopy [7] at $\lambda = 486$ nm. The transition frequency of this purely leptonic system was determined at a level of 10^{-8} and QED corrections which to a large fraction are due to virtual annihilation could be investigated. The electron and positron masses were found to be equal to 4×10^{-8}. However, the resolution of the measurement was limited by the frequency chirping effect due to the pulsed excitation, too.

Within the last decade many efforts were undertaken to circumvent the problems connected with the pulsed amplification by using cw light for exciting the transitions. In the case of hydrogen it was possible to generate sufficiently intense cw laser light using originally a sum frequency mixing technique [8] and later frequency doubling in β-Barium BOrate (BBO) [9] to improve the resolution of the frequency measurement considerably. For positronium the 1^3S_1–2^3S_1 energy interval was also remeasured using high power cw laser light in a high-finesse buildup cavity with an improved accuracy of 10^{-9} [10].

However, for many high-precision experiments the pulsed character of the laser light is essential. Recently, the 1^1S–2^1P energy interval in ^{4}He was measured for the first time by laser spectroscopy with a relative accuracy of 10^{-7} [11]. High-precision optical measurements on He are of great importance for basic atomic theory and recent

Dedicated to Prof. F.P. Schäfer on the occasion of his 65th birthday.

progress in the theoretical treatment of this fundamental three-body system allows to calculate its energy levels with unprecedented accuracy. In particular, a measurement of the 1^1S–2^1P energy interval provides a stringent test for two-electron contributions to the Lamb shift. Since there are no suitable laser sources available at the transition wavelength of 58.4 nm, the necessary radiation was produced by fifth-harmonic generation from a frequency-doubled, pulsed dye laser beam at 584 nm. The main uncertainty in this experiment was introduced by the frequency calibration at the fundamental wavelength. The experiment was followed by a more precise measurement using a cw dye laser in the visible, which combined the possibilities of a precise frequency calibration employing Doppler-free saturation spectroscopy and amplifying parts of the light in a pulsed dye laser amplifier for the harmonic generation [12]. In this way, the absolute frequency accuracy was improved to 3 parts in 10^8. Again the major contribution to the systematic errors of the measurement was caused by the chirping effect.

The work described here has been started in connection with an experiment which aims for a highly accurate determination of the $1S$–$2S$ transition frequency in the hydrogen-like muonium atom (μ^+e^-) [13], the Coulomb bound state of a positive muon (μ^+) and an electron (e^-). Muonium as a purely leptonic system consists of two particles which can be considered point-like down to dimensions of 10^{-18} m [14]. Its energy levels can be calculated to very high accuracy almost exclusively by QED. There are no complications arising from nuclear structure which are causing at present the main uncertainty in the theory of atomic hydrogen. In fact, today many spectroscopic experiments in hydrogen are more accurate than their theoretical description, because of the not well enough known charge distribution within the proton and the dynamics of its charge-carrying constituents. The muonium atom is not a particle–antiparticle system like positronium and hence no corrections for annihilations need to be accounted for when the level energies are calculated. Precise measurements of transition frequencies in muonium, in particular its hyperfine and $1S$–$2S$ splittings, can provide therefore sensitive experimental tests of the theoretical calculations within the framework of QED and furthermore yield precise values for fundamental constants [15].

An experiment to measure the $1S$–$2S$ energy splitting in muonium by Doppler-free two-photon spectroscopy is under way at the worldwide brightest pulsed muon channel at the ISIS synchrotron of the Rutherford Appleton Laboratory in Chilton, UK. For an efficient excitation of this transition a powerful laser system is indispensable. An accurate result can only be achieved with a high transition probability, because of the moderate muonium production rates of typically a few hundred atoms per second. A first measurement could already be performed in which the transition frequency was determined at a level of 2×10^{-8} [16] in agreement with the result of a pioneering experiment at KEK in Tsukuba, Japan [17]. The QED contributions beyond the level separation predicted in Dirac theory were tested at a level of 8×10^{-3} and an accurate value of the muon mass could be extracted. The optical part of the experiment consisted of a narrowband cw ring dye laser at $\lambda = 488$ nm which is amplified in a four stage, XeCl excimer pumped dye amplifier and frequency-doubled in a nonlinear β-BBO crystal to induce the $1S$–$2S$ two-photon resonance. The accuracy of the results was limited like in the foregoing examples by optical phase perturbations during the pumping process of the laser amplifier.

The experiments described below were carried out in order to understand better the systematic corrections necessary in the muonium $1S$–$2S$ experiment and to improve the accuracy of future frequency measurements which promise to yield most accurate values for the mass and the magnetic moment of the positive muon at a level of 10^{-7} which requires the center frequency of the resonance to be determined better than 10^{-9} relative accuracy.

1 Measurement of the phase perturbations with a heterodyne method

An XeCl excimer laser (Lambda Physik EMG 201) at a wavelength $\lambda = 308$ nm with a typical pulse duration of about 20 ns and pulse energies up to 0.5 J was used to optically pump Coumarin 102, 153 and 307 dyes in a commercial dye laser amplifier (Lambda Physik FL-2002EC). The time-dependent phase shifts which were introduced into the amplified light by rapid changes of the refractive index during the pump process were investigated in a heterodyne setup (Fig. 1). Measurements were carried out with seed beams from a single mode cw dye laser at wavelengths of $\lambda = 550$ nm or $\lambda = 488$ nm. In order to separate effects related to properties of the solvent used for the laser dyes from possible influences connected to the population inversion in the dye molecules [18] a single mode He–Ne laser beam at 633 nm was employed for sensing the phase perturbations in the dye cuevettes far off the resonances in the dye molecules which can be used for amplification. The red He–Ne laser light propagated collinearly with the green or blue laser seed beam on an identical optical path through the active volume in the dye cuevettes. Part of each of the cw beams was split off prior to the amplification region, was frequency-shifted by 150 MHz in an Acousto-Optic Modulator (AOM) (Isle Optics, Model LM210-1) and was overlaid with the respective perturbed light on a fast photodiode (Hamamatsu S2381). The resulting heterodyne beat note was recorded with a 1 GHz digitizing oscilloscope. The heterodyne signal $S(t)$ is given through

$$S(t) = A(t) \cdot \sin(2\pi\nu_{\mathrm{AOM}}t - \Phi(t)) + N, \tag{1}$$

where $A(t)$ is the time-dependent amplitude, ν_{AOM} the 150 MHz frequency offset, $\Phi(t)$ the time-dependent phase and N accounts for a constant background offset. In contrast to previous work [18, 19], which used distinctively different methods of analyzing the data, the function $S(t)$ was fitted section-wise in a time window containing six data points to the experimental signal with a least-squares method; a full set of parameters (A, Φ, N) was extracted for every time bin. The time derivative of the argument of the

sine function in Eq. (1) yields the "instantaneous" frequency [20]

$$\nu_{\text{inst}} = \nu_{\text{AOM}} - \frac{1}{2\pi}\frac{\mathrm{d}\Phi}{\mathrm{d}t}. \tag{2}$$

For the He–Ne laser light the 150 MHz beat signal which is observed prior to firing the excimer laser is affected by the intense pulsed pump light (Fig. 2). The perturbation of the optical phase $\Phi(t)$ extends over several ms far beyond the duration of the laser pulse. This matches the time scale of the exchange of the dye liquid in the active volume as it was constantly circulated through the cuevettes. The instantaneous frequency rises to a sharp peak of typically several 10 MHz above the frequency offset and falls off almost completely within the duration of the laser pulse. We define the chirp frequency ν_c as the maximum of the excursion of the instantaneous frequency with respect to the 150 MHz offset

$$\nu_c = -\frac{1}{2\pi}\left(\frac{\mathrm{d}\Phi(t)}{\mathrm{d}t}\right)_{\max}. \tag{3}$$

The instantaneous frequency varied under normal operation on a shot by shot basis typically by as much as 15% (rms).

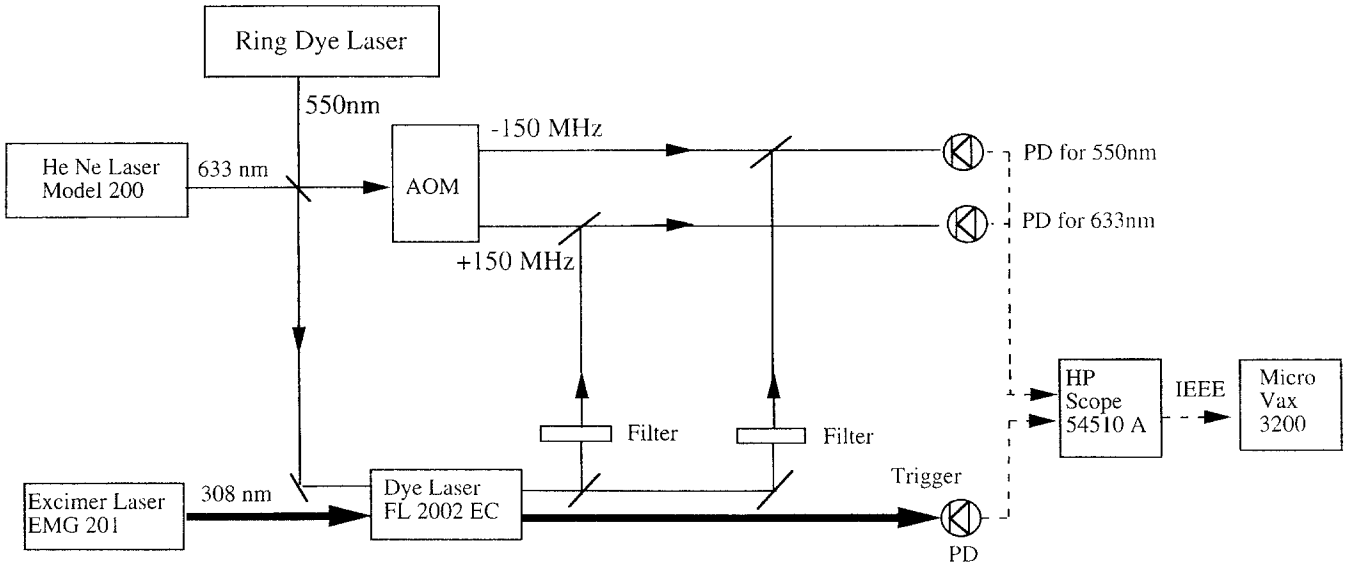

Fig. 1. A red He–Ne laser beam at 633 nm was copropagating with the green (or blue) cw seed laser beam at 550 nm (488 nm) on the same optical path in the dye cells. Part of the cw light was shifted in frequency by 150 MHz in an Acousto-Optic Modulator (AOM) and overlaid with the respective phase perturbed light on fast PhotoDiodes (PD) to obtain the heterodyne beat note

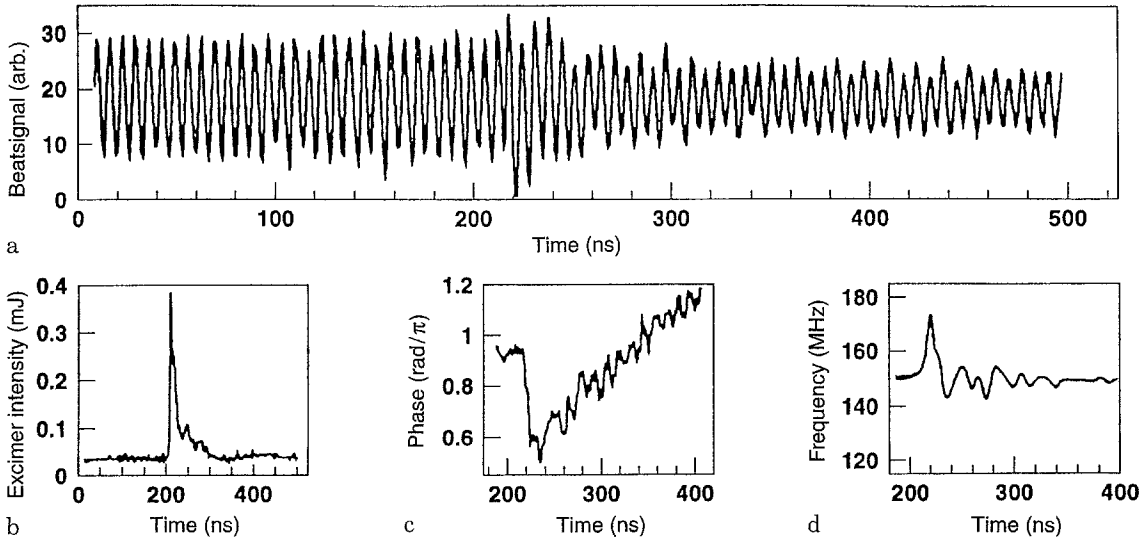

Fig. 2. The heterodyne signal (**a**) is affected by the excimer pump pulse (**b**). The evolution of the optical phase (**c**) and the corresponding frequency shift (**d**) are derived for each time bin of 1 ns width from a section wise numerical least-squares fit to a sine curve which included six neighboring experimental data points each

2 Dependence of the frequency chirp on different experimental conditions

The influences of various experimental parameters on the magnitude of the chirp frequency have been investigated. All observed chirping effects could be sensed by the He–Ne laser light. No additional chirping was found for the green or blue laser light which exceeded the effect measured for the red light within the accuracy of our results.

We have observed an influence of the concentrations of different dyes (Coumarin 102, 153, 307) in different chemicals as solvents (methanol, ethylene glycol, benzyl alcohol and water) on the frequency chirping only for very low dye concentrations below 0.1 g/l for all dye solutions. For higher concentrations the chirping effect appeared to be independent of this parameter. However, a strong correlation between the thermal expansivity of the used solvents and the magnitude of the measured chirp frequency was found (Fig. 3). This could be verified particularly when water was used as a solvent by varying the temperature between 20 and 70°C where the thermal expansivities are distinctively different. The chirp frequency depended linearly on the pump laser intensity in the range between 50 and 500 mJ. It decreased when the pump beam was defocussed. No direct correlation of the chirp frequency could be established with other parameters like the heat conductivity or the heat capacity of the dye solutions.

These observations led us to assume that the phase perturbations depend mostly on the deposition of energy in the active volume by the pump pulse and the resulting thermal expansion of the dye solvents. For our experimental conditions we could not determine any significant influence of the resonant behavior of the molecules or the inversion in the medium. The frequency shifts revealed with the heterodyne method correspond in their magnitude to earlier measurements obtained by a time averaging interferometric method which were carried out for Coumarin 102 and 307 at a wavelength $\lambda = 488$ nm in the course of the muonium 1*S*–2*S* measurements at the RAL [17].

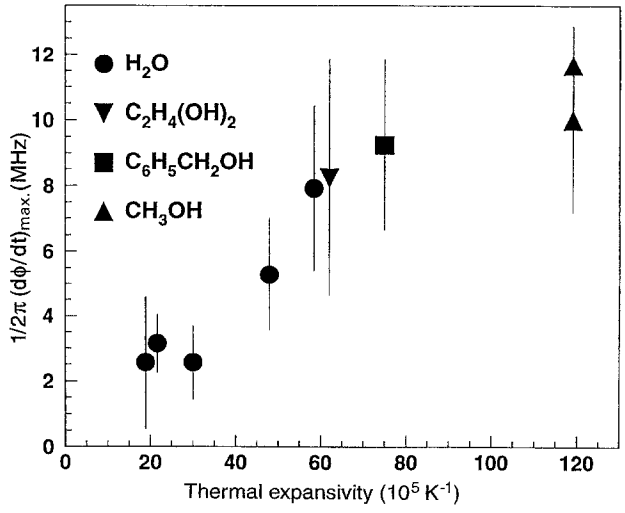

Fig. 3. Dependence of the chirp frequency on the thermal expansivity of different dye solvents. The measurements using water were performed at temperatures of 20, 21, 30, 53 and 70°C.

3 Compensation of frequency chirping with an electro-optic phase modulator

In order to compensate the pump-laser-induced frequency chirping and to test the analyzing method used to characterize the time-dependent behavior of the beat signal a fast Electro-Optic Modulator (EOM) was built. Since the relevant phase perturbations occur within nanoseconds, the device was designed in a traveling-wave configuration [21] to allow a manipulation of the laser light on the same timescale (Fig. 4). The EOM consists of two $1.0 \times 0.75 \times 25$ mm^3 lithium tantalate crystals ($LiTaO_3$). The crystals have been manufactured for an operation at 488 nm, the end surfaces are antireflection-coated and gold electrodes are deposited on the 1.0×25 mm^2 side faces to allow the application of a voltage parallel to the optical crystal axis. The ratio of height to width was selected for the crystals to appear as a 50 Ω transmission line [22]. Laser light and voltage pulses propagate at the same speed along the crystals. The externally applied electric field changes the extraordinary refractive index n_e. The phase $\Phi(t)$ of a light wave which is polarized parallel to the optical axis changes proportional to the applied voltage pulse $U(t)$ [23]:

$$\Delta\Phi(t) = \frac{\pi}{\lambda} n_e^3 r_{33} \frac{l}{d} U(t), \tag{4}$$

where λ is the wavelength, l is the length, d is the spacing of the crystal and r_{33} is the relevant electro-optic coefficient of the crystal. The half-wave voltage $U_{\lambda/2}$ which corresponds to a phase shift of π is 29 V at a wavelength of 488 nm. The frequency response of the modulator in the range of 300 MHz–1.3 GHz, which is depicted in Fig. 5, was determined by measuring the peak intensities of the optical sidebands generated from a single mode He–Ne laser beam passing through the device when it was driven by a sinusoidal voltage. The observed –3 dB bandwidth of 1.2 GHz is mostly due to the bandwidth limit of the driving amplifier used in this test.

The EOM was employed to verify the newly developed analyzing method for extracting the chirp frequency. Phase shifts were introduced into the light of a single mode He–Ne laser by applying voltage pulses of typically

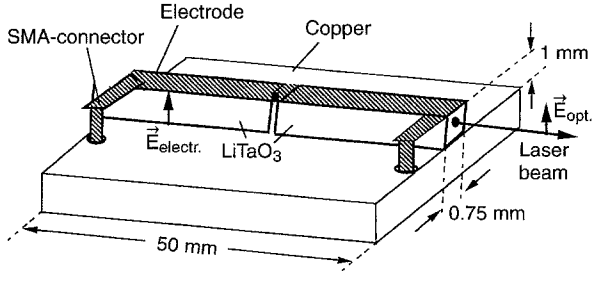

Fig. 4. The EOM in traveling-wave configuration. The laser light and a voltage pulse are propagating with the same speed along the electro-optic crystals.

10 V_{PP} which had rise times of a few ns and widths ranging from 10 to 30 ns to the device. The electrical waveforms reconstructed from the measured optical phase shifts were in good agreement with the actually applied voltage pulses (Fig. 6).

When step-function-voltage pulses (Fig. 7) with rise times of the order of 10 ns were applied to the EOM, artificial phase shifts could be generated similar to the phase disturbance observed in the amplified light during the duration of an excimer pump laser pulse. The magnitude of the resulting chirp frequency depended for a fixed rise time on the voltage amplitude of the step and amounted for 10 V to about 10 MHz. When such voltage pulses were applied to the EOM, the pump-laser-induced chirp frequencies could be lowered by a factor up to 4, if the sign, amplitude, rise time and timing were adjusted appropriately. Because of the shot-to-shot variations in the chirp frequency, a compensation significantly beyond that level appears not possible unless an active feedback can be established, which would require the pulsed light to be stored in an optical delay line while the necessary electronical pulse is generated to be applied to the EOM.

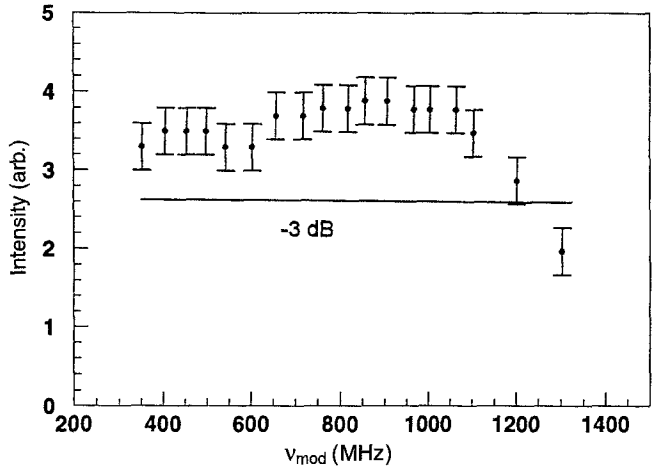

Fig. 5. The intensity of the first-order sidebands generated by electro-optic modulation from a single mode He–Ne laser beam for constant modulation power versus the modulation frequency. The observed bandwidth of the system is about 1.2 GHz

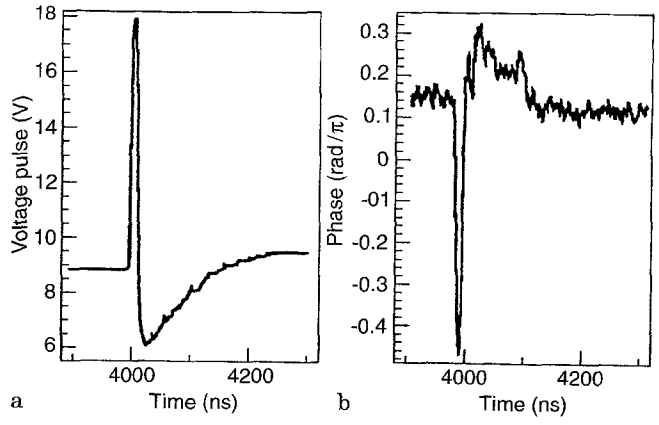

Fig. 6. The analysis procedures for the beat signal were tested by applying a voltage pulses (**a**) to the modulator. The measured optical phase change (**b**) corresponds in size and form (opposite sign) to the original voltage pulse.

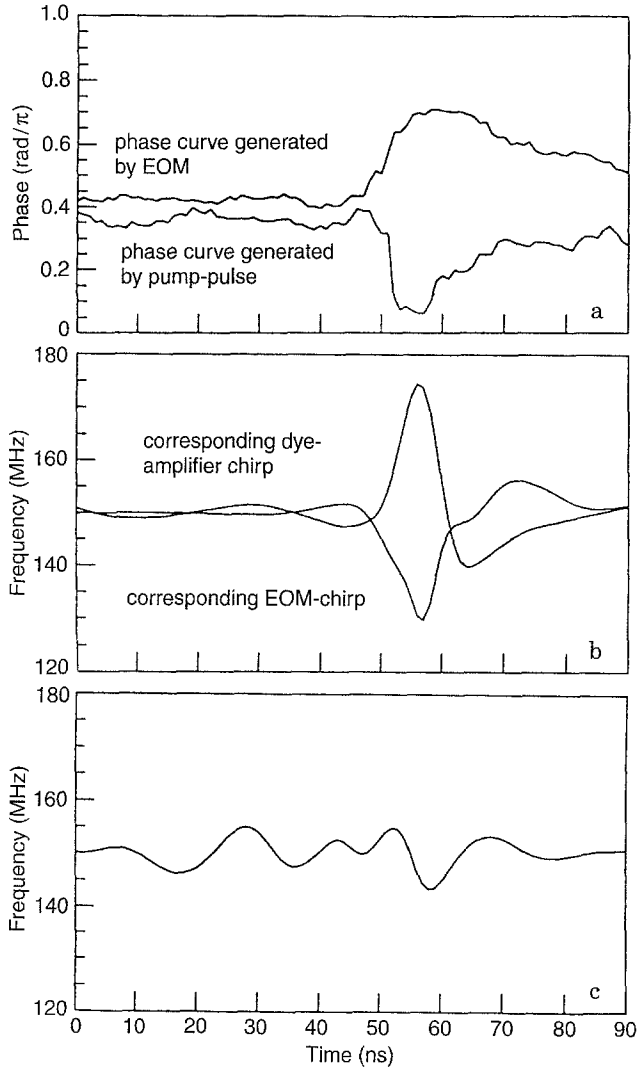

Fig. 7. By applying an appropriately shaped voltage (**a**) pulse to the EOM the artificially generated phase shifts could be made similar to the phase perturbations introduced in the course of the pumping process into the amplified light. The calculated chirp frequencies have similar magnitude but opposite signs (**b**). The phase shifts in an amplified laser beam could be partly compensated when it passed through an EOM to which a properly shaped and timed voltage pulse was applied and the chirp frequency could be significantly reduced (**c**)

4 Conclusion

An optical heterodyne setup was employed to investigate fast phase perturbations of laser light due to changes of the index of refraction in an excimer laser pumped dye laser amplifier. A new method has been developed for analyzing the heterodyne response. For the conditions of this experiment the observed changes in the instantaneous frequency depend mostly on the thermal expansivity of

the dye solutions and no significant dependence on the properties of the dye molecules and the population inversion was found as it was reported for other measurements where solvent related effects were not separated in the experimental approach [18].

A traveling wave EOM with a −3 dB bandwidth above 1.2 GHz was built which allowed to verify the analysis procedure by introducing artificially phase perturbations to the laser light. The magnitude of the chirp frequency originating from the pulsed pumping light could be reduced to a quarter of its previous value by applying an appropriately shaped and timed voltage pulse to the EOM. The latter aspect is of particular interest, because the frequency chirping not only introduces a systematic frequency offset, which reduces the accuracy of the frequency calibration, but it also causes in the case of transitions which are narrower than the laser bandwidth a reduction of the transition rate like, for example, in the $1S$–$2S$ transition in muonium.

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