

Chirp-corrected, nanosecond Ti:sapphire laser with 6 MHz linewidth for spectroscopy of antiprotonic helium

Masaki Hori^{1,2,*} and Andreas Dax²

¹Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany

²Department of Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

*Corresponding author: masaki.hori@mpq.mpg.de

Received December 8, 2008; revised February 24, 2009; accepted February 27, 2009;
 posted March 13, 2009 (Doc. ID 105070); published April 14, 2009

A nanosecond titanium sapphire laser with spectral linewidth $\Gamma_{\text{pl}} \sim 6$ MHz and pulse energy of 50–100 mJ was demonstrated by using an intracavity electro-optic modulator to correct the frequency chirp in the output beam. The laser was referenced against a femtosecond frequency comb and used to measure the 6s–8s ($F=4$) two-photon transition frequency of Cs with a precision of 1.4 parts in 10^9 . © 2009 Optical Society of America

OCIS codes: 300.6360, 140.3520, 140.3590, 140.3538, 120.5060.

Nanosecond lasers with narrow (<100 MHz) linewidth have been used in many high-precision spectroscopy experiments, e.g., measuring UV lines in He, H₂, Mg, and exotic atoms [1–5]. These experiments determined the Lamb shift in He and muonium [1,4], limits on the possible variation of the fine structure constant over Gy-scale periods [2,3], and the antiproton-to-electron mass ratio [5]. While cw lasers and frequency combs can in principle achieve much higher spectral resolutions, so far these experiments have only been carried out using the high ($E > 10$ –100 mJ) output energies available from nanosecond lasers. The latter lasers typically pulse amplify a cw seed beam with a large gain of 10^6 – 10^9 . We might naively expect that the frequency of the amplified light ν_{pl} would then be equal to the seed frequency ν_{cw} and that the linewidth Γ_{pl} would be limited only by the Fourier transform limit corresponding to the laser pulse length Δt . Actual lasers, however, are modulated by nanosecond-scale changes in the refractive index n_c of the gain media during the amplification. This causes a frequency chirp,

$$\Delta\nu_c = \nu_{\text{pl}} - \nu_{\text{cw}} = -\frac{1}{2\pi} \frac{d\phi}{dt} \propto \frac{dn_c}{dt}, \quad (1)$$

wherein ϕ denotes the optical phase. In amplifiers employing cavities, so-called “mode-pulling” [3,4,6] effects can cause additional shifts in ν_{pl} , as changes in n_c detune the cavity from the seed frequency ν_{cw} during buildup of the laser pulse. All these effects can broaden the laser linewidth and shift its frequency by several tens of megahertz or more, especially in high-power lasers involving large values of dn_c/dt . It is therefore vital in any spectroscopy experiment to characterize $\Delta\nu_c$.

Fee *et al.* [7] introduced a heterodyne technique to measure the time evolution of $\Delta\nu_c$. Two groups [1,4] then used an electro-optic modulator (EOM) to apply an “antichirp” to a dye laser, thereby canceling $\Delta\nu_c$. This technique was later applied to a flashlamp-pumped alexandrite laser [4,8] but not, to our knowledge, to titanium sapphire (Ti:S) lasers pumped by

solid-state lasers [9,10]. A nanosecond optical parametric oscillator with $E \sim 5$ μJ and small $\Delta\nu_c$ values [6] was recently used to measure the $F=4$ hyperfine component in the 6s_{1/2}–8s_{1/2} two-photon transition in Cs. An FWHM linewidth $\Gamma_{\text{Cs}} \sim 18$ MHz was reported (all linewidths quoted in this paper are at the laser wavelength $\lambda = 822$ nm). We here used an intracavity EOM to correct the chirp in a Ti:sapphire (Ti:S) laser of energy $E = 50$ –100 mJ, thereby achieving a resolution of $\Gamma < 5$ MHz on this Cs line. We plan to use this laser in experiments of antiprotonic helium [5], and so it was vital to study its precision against a known Cs frequency.

Our Ti:S ring laser (circumference of 800 mm) generated 40–100-ns-long laser pulses of energy $E \sim 8$ –15 mJ. This beam was amplified to $E \sim 50$ –100 mJ by making three passes through another Ti:S crystal (indicated by C2 in Fig. 1). The cavity consisted of (i) a concave mirror (M1) with a radius of curvature $R_c = 4$ m mounted on a piezo element, (ii) a flat output coupler (OC) of reflectivity $R = 80\%$ – 90% , and (iii) a dispersive prism [9] that

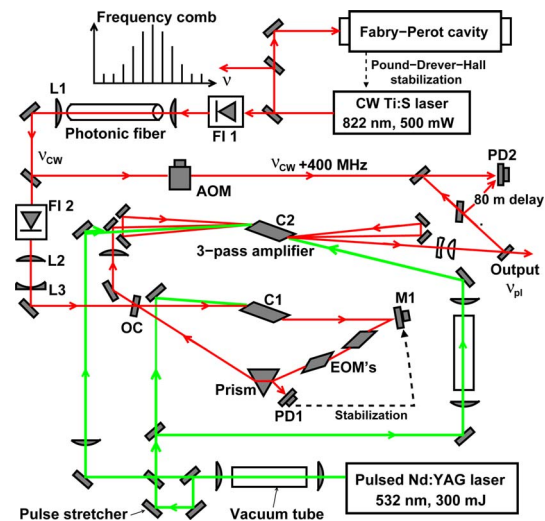


Fig. 1. (Color online) Layout of the chirp-compensated Ti:S laser; abbreviations are defined in the text.

coarsely determined the laser wavelength. A 20-mm-long Ti:S crystal (C1—identical to C2) cut at Brewster's angle with its c axis parallel to the laser polarization was placed between OC and M1. The crystal had an absorption of $\alpha_{514}=2\text{ cm}^{-1}$ at $\lambda=514\text{ nm}$. Its high figure of merit $\alpha_{514}/\alpha_{820}\sim 300$ prevented any secondary chirp caused by IR light being reabsorbed by crystal C1. A pair of EOM crystals made of potassium dideuterium phosphate cut at Brewster's angle was placed between OC and the prism. The laser was operated at wavelengths $\lambda=726\text{--}941\text{ nm}$.

Among the advantages of this design for minimizing Γ_{pl} were that, according to Eq. (1), the chirp is small for long laser pulses with smooth temporal profiles involving small gradients dn_c/dt . The cavity was therefore designed for a large transverse mode diameter $d_t\sim 1.1\text{ mm}$ of the circulating beam. This allowed us to reduce the power density in the Ti:S crystal and thus its single-pass gain, which contributed to the generation of long laser pulses. This also supported high intracavity laser powers without damaging the EOMs and mirrors. Furthermore, most of the light gain ($>10^6$) occurred in the laser cavity, where any chirp induced by a pass through crystal C1 could be readily corrected by a corresponding pass through the EOMs. A single EOM pass induced a frequency shift $\Delta\nu_e(t)$, which was roughly proportional to the differential $dV(t)/dt$ of the voltage waveform applied to its electrodes. The circulating beam accumulated such shifts over many passes, i.e.,

$$\Delta\nu_e(t) \propto \sum_{i=1} \frac{dV(t_i)}{dt} \sim \int_{t=0}^{\Delta t} \frac{dV(t)}{dt} dt \propto V(t), \quad (2)$$

so that $\Delta\nu_e(t)$ now became proportional to $V(t)$. The condition $\Delta\nu_c(t)=-\Delta\nu_e(t)$ needed to correct the chirp could thus be achieved at lower voltages $V(t)$, compared to the single-pass EOM geometry used in the dye laser case [1,5] wherein $\Delta\nu_e \propto dV/dt$.

The Nd:YAG pump laser produced 3-ns-long laser pulses with $\lambda=532\text{ nm}$ and $E=300\text{ mJ}$. Its injection-seeded phase-conjugate mirror cavity allowed it to fire with a low energy fluctuation of 5% and repetition rate $f\sim 0.1\text{ Hz}$. To prevent this high peak power from damaging the Ti:S crystals the beam was first stretched to $\Delta t\sim 10\text{ ns}$ using some beam splitters [5]. A 20–40 mJ portion of this beam was then focused with a diameter of 1.2 mm onto crystal C1 using a lens of focal length $f_c=1000\text{ mm}$. Two beams of $E\sim 130$ and 90 mJ likewise pumped crystal C2 from both ends. The chirp critically depended on this alignment. To minimize any long-term drift in their positions, all beams were relay imaged onto the crystals using 600-mm-long vacuum tubes with lenses of $f_c=400$ and 300 mm on their ends.

The seed beam was provided by a cw Ti:S laser stabilized by the Pound–Drever–Hall technique to a Fabry–Pérot cavity. The cavity had a free spectral range of 390 MHz and finesse of 2000, and it was thermally stabilized and suspended in a vacuum chamber. The laser linewidth and frequency drift were $\Gamma\leq 100\text{ kHz}$ and $\sim 100\text{ kHz/h}$. Its frequency ν_{cw}

was measured with a precision of $\sim 100\text{ kHz}$ by a Ti:S femtosecond comb (Menlo Systems FC8004) referenced to a global-positioning-system-disciplined quartz oscillator.

The seed beam traversed two Faraday isolators (FI1 and FI2), a single-mode fiber, and an aspheric lens (L1) of $f_c=8\text{ mm}$. Lenses L2 and L3 then coupled the p -polarized beam of power $P=50\text{ mW}$ into the pulsed Ti:S cavity through mirror OC. The cavity was locked to the seed beam to maximize the power circulating in it by (i) dithering the piezo-mounted mirror M1 at frequency $f_d\sim 26\text{ kHz}$, (ii) measuring the amplitude-modulated light reflected from the prism onto the photodiode PD1, (iii) a correction signal from the diode was generated using a lock-in amplifier referenced to f_d . This was fed back to the piezo. To prevent the strong pulsed light from disturbing this lock, a track-and-hold regulator froze the correction signal $\sim 100\text{ }\mu\text{s}$ before the pumping.

The laser pulse was attenuated to $E=1\text{--}12\text{ }\mu\text{J}$ to carry out the Cs spectroscopy. A collimated beam of diameter $d=1.5\text{ mm}$ passed through a pinhole before entering a 100-mm-long Pyrex cell filled with Cs. The beam emerging from the other side traversed a second pinhole, retroreflected off a mirror, and reentered the cell, overlapped with the counterpropagating beam. Some of the Cs excited to the $8s$ state decayed via the transition chain $8s\rightarrow 7p\rightarrow 6s$. The 456 nm light emerging from this was detected by a photomultiplier with a blue filter. The cell was heated to $T=60\text{--}120^\circ\text{C}$ with a stability $\pm 0.1^\circ\text{C}$. It was magnetically shielded in a mu-metal box.

The time evolution of the chirp $\Delta\nu_c$ was measured by first diverting a 2 mW part of the seed beam into an acousto-optic modulator that shifted its frequency by +400 MHz. This cw beam was then superimposed with the pulsed beam on photodiode PD2, and the resulting heterodyne signal $S_H(t)$ was recorded by a digital oscilloscope of analog bandwidth and sampling rate $f=3$ and 10 GHz. Part of the laser pulse propagated along an 80-m-long delay line before striking an opaque diffuser placed near PD2. The photodiode thus measured both $S_H(t)$ and the carrier envelope $S_E(t)$. Fourier analysis was used to obtain $\Delta\nu_c(t)$ from these two signals. Simulations [1,3] showed that a precision of $<0.4\text{ MHz}$ on $\Delta\nu_c(t)$ could be achieved over most of the 100-ns-long laser pulse.

The uncorrected chirp [Fig. 2(b), dashed curve] shifted to $\Delta\nu_c(t)<-10\text{ MHz}$ before returning to zero during the laser pulse. The observed Cs line [Fig. 2(c)] was correspondingly asymmetric and its centroid shifted by -10 MHz relative to ν_{cw} , whereas its linewidth $\Gamma_{\text{Cs}}\sim 15\text{ MHz}$ was much larger than the Fourier limit of $S_E(t)$. At high pump energies $E>40\text{ mJ}$ and small focus $d_t\leq 1\text{ mm}$ in C1, the cavity mode-pulling became so large that two longitudinal modes were amplified, causing beat notes to appear in $S_E(t)$. The laser pulse shortened to $\Delta t<40\text{ ns}$, whereas its energy and timing jitter increased.

The integrating effect of multiple EOM passes described by Eq. (2) is illustrated in Fig. 3(a). Stepwise high-voltage pulses of 0.55 and -1.3 kV were applied

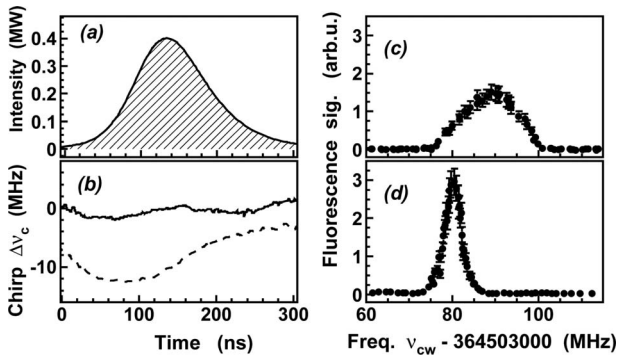


Fig. 2. Time evolution of the (a) intensity and (b) frequency chirp of Ti:S laser pulses with (solid curve) and without (dashed curve) chirp compensation. The $6s-8s$ ($F=4$) two-photon resonance of Cs as a function of ν_{cw} (c) without and (d) with compensation.

to the EOMs at $t_1=25$ and $t_2=45$ ns. As expected, $\Delta\nu_c$ was proportional to $V(t)$ and shifted by $+50$ MHz and then -120 MHz within a single laser pulse, following a rectangular function. The Cs spectrum measured using this laser pulse shows three peaks at the expected positions [Fig. 3(b)]. We corrected the chirp to $|\Delta\nu_c(t)| \leq 2$ MHz over most of the laser pulse [solid curve in Fig. 2(b)] by using a network of field-effect transistor switches to iteratively adjust the amplitude (typically 50–150 V) and waveform of $V(t)$. The gradients $|dV/dt| = 1-30$ V/ns were adjusted by tuning some load resistors and capacitors connected to the EOMs. The above problem of multimode oscillation was eliminated, whereas fluctuations in the energy ($\sim 10\%$) and timing (± 10 ns) were reduced. The laser linewidth is now inferred [10] to be nearly equal to the Fourier limit $\Gamma_{pl} \sim 6$ MHz corresponding to the temporal profile $S_E(t)$ of Fig. 2(a). The observed Cs resonance [Fig. 2(d)] is three times narrower ($\Gamma_{Cs} \sim 4.5$ MHz) than in the uncorrected case, whereas its centroid is $\Delta\nu_c \sim 0$. This two-photon linewidth is expected to decrease [11] by a ratio $\Gamma_{Cs}/\Gamma_{pl} \sim 0.7$ when the transition is driven by an ideal Fourier-limited laser pulse with a Gaussian temporal profile, and indeed we here see this narrowing.

We calculated the expected Cs line shape by integrating two-photon optical rate equations. The laser frequency ν_{pl} was here modulated according to its measured time evolution. We included effects due to the temporal and spatial profiles of the pulse. By fitting the data with this theoretical curve, we deter-

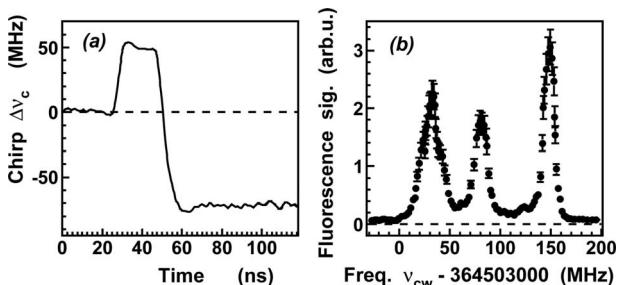


Fig. 3. (a) Chirp induced by applying two voltage pulses at $t_1=25$ and $t_2=45$ ns to the EOM, and (b) the resulting $6s-8s$ ($F=4$) Cs spectrum as a function of ν_{cw} ; see text.

mined the resonance centroid as $\nu_{6s-8s}(F=4) = 364,503,080.3(5)$ MHz. This is within 100 kHz of published values [12] of much higher precision. We estimated the effect of the measurement error of the chirp $\Delta\nu_c(t)$ by varying its value used in the above derivation. This resulted in an uncertainty of 0.3 MHz on ν_{6s-8s} . Other sources of error are the statistical one due to the fit (0.3 MHz) and the accuracy on ν_{cw} (0.1 MHz) measured by the frequency comb. Measurements at laser energies of $1-12 \mu\text{J}$ showed ac Stark shifts between -0.3 and -3 MHz. This agrees with previous experimental values -0.2 Hz/(mW/cm²) [12]. We extrapolated the results to zero power to determine ν_{6s-8s} ; the error associated with this procedure was 0.3 MHz. The collisional shift is small ~ 10 kHz/mTorr [12]; indeed, measurements at cell temperatures between $T=60^\circ\text{C}$ and 100°C showed no significant shift. Magnetic shifts are < 10 kHz [12]. All these errors were quadratically added.

We conclude that the EOM chirp correction technique has resulted in one of the smallest linewidth (6 MHz) reported so far for a Ti:S laser of output energy $E=50-100$ mJ. Two-photon spectroscopy of Cs indicated that the laser can be used in experiments at parts-per-billion scale precisions, such as those planned for antiprotonic helium.

We are indebted to the ASACUSA collaboration, K. Jungmann, and Th. Udem. This work was supported by the European Young Investigator award of the European Science Foundation, the Munich-Centre for Advanced Photonics of Deutsche Forschungsgemeinschaft, and Monbukagakusho.

References

1. K. S. E. Eikema, W. Ubachs, W. Vassen, and W. Hogervorst, *Phys. Rev. A* **55**, 1866 (1997).
2. A. Yiannopoulou, N. Melikechi, S. Gangopadhyay, J. C. Meiners, C. H. Cheng, and E. E. Eyler, *Phys. Rev. A* **73**, 022506 (2006).
3. S. Hannemann, E. J. Salumbides, S. Witte, R. Th. Zinkstok, E.-J. van Duijn, K. S. E. Eikema, and W. Ubachs, *Phys. Rev. A* **74**, 012505 (2006).
4. V. Meyer and HORSSY collaboration, *Phys. Rev. Lett.* **84** 1136 (2000), and references therein.
5. M. Hori, A. Dax, J. Eades, K. Gomikawa, R. S. Hayano, N. Ono, W. Pirkl, E. Widmann, H. A. Torii, B. Juhász, D. Barna, and D. Horváth, *Phys. Rev. Lett.* **96**, 243401 (2006).
6. M. Kono, K. G. H. Baldwin, Y. He, R. T. White, and B. J. Orr, *Opt. Lett.* **30**, 3413 (2005).
7. M. S. Fee, K. Danzmann, and S. Chu, *Phys. Rev. A* **45**, 4911 (1992).
8. R. Nicolaescu, T. Walther, and E.S. Fry, in *Lasers and Electro-Optics 1998 Technical Digest* (Optical Society of America, 1998), paper CTuM69.
9. T. Walther, M. P. Larsen, and E. S. Fry, *Appl. Opt.* **40**, 3046 (2001).
10. K. F. Wall, P. A. Schulz, R. L. Aggarwal, P. Lacovara, and A. Sanchez, *IEEE J. Quantum Electron.* **29**, 1505 (1993).
11. F. S. Cataliotti, R. Scheunemann, T. W. Hänsch, and M. Weitz, *Phys. Rev. Lett.* **87**, 113601 (2001).
12. P. Fendel, S. D. Bergeson, Th. Udem, and T. W. Hänsch, *Opt. Lett.* **32**, 701 (2007).