Ultrashort pulse Cr⁴⁺:YAG laser for high precision infrared frequency interval measurements

A. J. Alcock, P. Ma, and P. J. Poole
Institute for Microstructural Sciences, National Research Council of Canada, 1200 Montreal Road, Ottawa, Ontario K1A 0R6, Canada

S. Chepurov
Institute of Laser Physics, Russian Academy of Sciences, Novosibirsk, Russia

A. Czajkowski
Department of Physics, University of Ottawa. 150 Louis Pasteur, Ottawa, Ontario K1N 6N5 Canada

J. E. Bernard and A. A. Madej
Institute for National Measurement Standards, National Research Council of Canada, 1200 Montreal Road, Ottawa, Ontario K1A 0R6 Canada

J. M. Fraser
Department of Physics, Queens University, Kingston, Ontario, K7L 3N6 Canada

I. V. Mitchell
Department of Physics and Astronomy, University of Western Ontario, London, Ontario N6A 3K7, Canada

I. T. Sorokina and E. Sorokin
Institut für Photonik, Technische Universität Wien, Gusshausstrasse 27/387, A1040 Wien, Austria

Abstract

A cavity stabilized, SESAM mode-locked Cr⁴⁺:YAG laser capable of generating sub-100 fs pulses has been developed. Locking the 130-MHz pulse repetition frequency to that of a hydrogen maser-referenced frequency synthesizer provides a 30-nm wide frequency comb for the 1530-nm wavelength region. In conjunction with a pair of acetylene stabilized, external cavity diode lasers, this laser provides a high precision measurement tool for the determination of acetylene transition frequencies.

1. Introduction

Although a mode-locked laser was first used for high-resolution spectroscopy in 1978 [1] it was not until 1999 that the application of ultrashort pulse lasers to precision optical frequency measurement entered a period of extremely rapid development [2-5]. These frequency comb systems have now opened new avenues in basic and applied domains of optical research and in the control of electro-magnetic radiation. In many cases, both the laser pulse repetition frequency (f_rep) and the carrier envelope offset (CEO) frequency are...
actively stabilized, and a broadband comb of precisely known optical frequencies is thus provided to measure the exact frequency of a particular source. Nevertheless, the simpler original version of this technique, which does not involve stabilization of the CEO frequency, allows the comb to be used as an extremely precise ruler for the measurement of an optical frequency interval. Provided that an appropriate reference source is available within the comb spectrum, the frequency of a second, uncalibrated source can be determined. To date, most of the research using combs has been performed using Ti:Sapphire mode-locked laser systems centered around 800 nm. Recently, mode-locked laser systems in the infrared [6,7] have been explored for operation near the infrared optical telecommunications bands at 1.5 and 1.3 \( \mu \text{m} \). These sources have an impact on telecommunication technology and associated applications in the near infrared.

In this paper, we describe a passively mode-locked Cr\(^{4+}\):YAG laser that is being used to measure the frequencies of acetylene overtone transitions that fall within the 1520 to 1553 nm wavelength range [8,9]. These lines will form a grid of known reference frequencies for the 1.5 \( \mu \text{m} \) telecommunication band and are the current internationally recognized optical frequency tie points in this region of the spectrum [10]. By stabilizing the pulse repetition rate of the CrYAG laser, the resulting comb of equally spaced optical frequencies can be used to determine the frequency difference between two external cavity diode lasers that are locked to different saturated absorption transitions of acetylene. One of the diode lasers is always locked to the P(16) line of \(^{13}\text{C}_2\text{H}_2\), which has been accepted as an international standard. An absolute frequency measurement, based on a Ti:Sapphire comb, has been reported recently [9]. The other laser system is then locked to a different transition for which the frequency is to be measured.

2. Description of the Cr\(^{4+}\):YAG laser

As shown in Fig. 1, the Cr\(^{4+}\):YAG laser is of conventional design with a Z-fold optical resonator and 10-cm radius-of-curvature concave mirrors at 32° folding angle to compensate the astigmatism of the Brewster-cut crystal. The arms of the symmetric resonator are 56 cm long leading to a pulse repetition frequency of 130 MHz. The 20-mm long, by 3-mm diameter Cr:YAG crystal is mounted in a copper holder which is maintained at a temperature of 20 C by means of a thermoelectric cooler. The pump source is a linearly polarized, 10-W ytterbium fiber laser, and a 10-cm focal length lens is used to focus the 1064-nm pump radiation within the crystal. For stability, and to reduce air currents within the optical resonator, the entire laser system is mounted on a single optical breadboard and placed within a Plexiglas enclosure.

Extremely stable mode-locked operation of the laser was obtained by means of a semiconductor saturable absorber mirror (SESAM) [11,12], which is mounted at one end of the optical resonator. An output coupler with a transmission of 0.5% terminated the other end of the resonator. Three, high reflectivity chirped mirrors, similar to those described in Ref [13] provide compensation for the group velocity dispersion of the CrYAG crystal. When a fourth chirped mirror is used to replace the SESAM and the 10-cm radius of curvature mirror located in front of it, Kerr lens mode-locking (KLM) of the laser can be obtained. Near-transform limited pulses of ~50 fs duration, at a center wavelength of 1560 nm, are observed (Fig. 2(a) and 2(b)). This mode of operation provides an output power of 200 mW but requires extremely careful alignment of the optical resonator, and is not sufficiently stable to provide reliable mode-locking during the several hours required to carry out an extensive series of frequency measurements. The problems associated with KLM operation are greatly reduced by incorporating a SESAM to control the mode-locked operation of the laser.
The SESAM used was similar to a hybrid device reported previously [14] in which a single In$_{0.53}$Ga$_{0.47}$As quantum well (Q.W.), giving a photo-luminescence peak at 1514 nm, provides the saturable absorption needed to modulate the reflectivity of a broadband, high reflectivity dielectric mirror. The hybrid-SESAM was fabricated in two steps, the first being the growth by means of chemical beam epitaxy (CBE) of the semiconductor layers on an InP wafer followed by the sputter deposition of a Ta$_2$O$_5$/SiO$_2$ quarter wave stack onto the top layer, see Fig. 3(a). In the second step, this structure was cemented onto a thin sapphire window and the InP wafer and 17-nm thick InP buffer layer were removed by using a combination of mechanical grinding and chemical etching. A second stage of etching was then used to remove the 150-nm InGaAs layer leaving only an extremely thin (3/4 λ) layer of semiconductor material, in which the 6-nm thick quantum well was embedded, followed by the multilayer dielectric reflector, see Fig. 3(b).

At this stage, insertion of the structure within the optical resonator of the Cr$^{4+}$:YAG laser resulted in stable mode-locked operation. However, as reported in Ref [14], the observed pulse duration was close to 0.5 ps. Pulse probe measurements of the SESAM's dynamic response were then carried out using 150 fs pulses from an optical parametric oscillator that could be tuned to the wavelength of the excitonic absorption. As shown in Fig. 4(a), a rapid change of reflectivity occurred at an incident fluence >2 μJ/cm$^2$. However, the subsequent recovery had a rapid component (sub-picosecond time constant) followed by a much slower component with a decay time of ~100 ps. The dynamic response of the SESAM was therefore modified by ion implantation [15] at the University of Western Ontario Tandetron accelerator. Fig. 4(b) shows that a much faster recovery time was observed after the SESAM was implanted with 500 keV phosphorus ions at a fluence of 10$^{12}$/cm$^2$. The implanted SESAM was not annealed and has been used to mode-lock the laser on a regular basis for more than one year.

As shown in Fig. 2(c) and 2(d), use of the ion-implanted SESAM resulted in the generation of mode-locked pulses of ~80 fs duration at a center wavelength of 1530 nm. Although the pulse duration was 50% longer than that obtained with KLM, and the average power was reduced to 80 mW, these penalties were completely outweighed by the robustness of the mode locking, which was maintained over many hours of operation. As can be seen from Fig. 2(b) and 2(d), the laser spectrum showed significant modulation for both KLM and SESAM operation. This appears to be associated with the presence of weak satellite pulses separated by several picoseconds from the main ultrashort pulse [13]. Although pump power, output coupling and alignment of the optical resonator influenced the number and strength of these satellites it was not possible to eliminate them completely when reliable mode-locking was obtained. However, their presence did not prevent the mode-locked frequency comb from being used to obtain highly accurate frequency interval measurements.

To provide a comb of precisely spaced optical resonator modes, the pulse repetition frequency ($f_{\text{rep}}$) of the mode-locked pulse train was stabilized by mounting the SESAM on a piezo-electric translator (PZT) and using an electronic servo-loop to lock $f_{\text{rep}}$ to the signal from a frequency synthesizer. The mechanical resonances of the SESAM-PZT structure limited the useful bandwidth of the servo loop. The open-loop unity gain frequency was 1 kHz and this was sufficient to achieve stable phase-locked operation of the laser to the reference. A 10 MHz signal derived from a NRC hydrogen maser was used as an external reference for the synthesizer. The NRC maser provides the needed short and medium term (T < 10$^5$ s) stability for the precise measurement of a frequency interval. Fractional stabilities of the maser are 3 × 10$^{-13}$ at 1 s improving to 2 × 10$^{-15}$ at 1000 s averaging time. The absolute maser frequency is provided via calibration against the NRC ensemble of primary Cs atomic time standards.
3. Frequency interval measurements

As shown in Fig. 5, the radiation from the Cr:YAG laser was split into two equal power beams that were combined with the radiation from a pair of external cavity diode lasers. These lasers have been developed as optical frequency standards that are stabilized to the saturation absorption resonances of the weak, $\nu_1 + \nu_3$ overtone transitions in $C_2H_2$. A more complete description of the apparatus is given in Refs. [8, 9]. As mentioned in the introduction, one of the diode lasers was always locked to the acetylene P(16) line while the other was locked to a line for which the frequency was to be determined. To select parts of the frequency comb close to the diode laser wavelengths, the combined Cr:YAG/diode laser beams were then incident on a pair of 600 line/mm diffraction gratings and the relevant portion of diffracted radiation focused onto two InGaAs photodetectors. Although the beat signals had sufficient power to be counted, they showed rapid frequency fluctuations and attempts to measure their frequencies directly with frequency counters were not successful. A difference frequency technique was therefore used to cancel out common-mode noise and the CEO frequency. RF bandpass filters were used to select one beat frequency near 340 MHz and a second in the 700-800 MHz range. The beat signals between the diode lasers and the comb were then amplified and the selected beats were further filtered and then mixed to yield a signal that was considerably less noisy than that of the individual beats. Finally, a tracking oscillator was locked to the mixer output and a frequency counter used to measure its frequency.

The frequency difference between the unknown line frequency and P(16) is given by

$$\Delta f = (m-n) f_{\text{rep}} \pm f_t$$

where “m-n” is an integer and $f_{\text{rep}}$ and $f_t$ are the laser repetition frequency and tracking oscillator frequency, respectively. Since the frequencies of the transitions in the $\nu_1 + \nu_3$ overtone band of $^{13}C_2H_2$ have previously been determined to an accuracy of ~1 MHz or less [16, 17], the value of the integer “m-n” and the sign can be readily determined.

In an initial experiment to demonstrate the application of the Cr:YAG comb, the difference in frequency between the $^{13}C_2H_2$ P(16) line and five other transitions (P(10), P(11), P(15), P(20) and P(21)) was measured. The absolute frequency measurements of these lines have been reported previously in Ref [9]. The frequency differences obtained with the Cr:YAG comb, the resulting line frequencies and those obtained from Ref. [9] are presented in Table I. It should be noted that in generating the results presented in Table I the frequency of the P(16) reference laser used in this study was 194 369 569 382 500 ± 1350 Hz (1$. The value of the integer “m-n” and the sign can be readily determined.

4. Summary

A cavity stabilized, passively mode-locked $Cr^{4+}:YAG$ laser has been used to generate a comb of precisely spaced optical frequencies in the 1530-nm wavelength region. With a bandwidth of ~ 4 THz this comb provides complete coverage of the $\nu_1 + \nu_3$ band of acetylene and thus offers an advantage over the electro-optic modulator (EOM)-based...
combs [17] and harmonic generation schemes [9,16] used in previous work. It has been used recently to obtain a complete atlas of 60 transitions in the $\nu_1+\nu_3$ band of $^{13}\text{C}_2\text{H}_2$ [18].

Acknowledgments

We would like to acknowledge the excellent technical support provided by Mr. Raymond Pelletier in the design and refinement of a number of the electronic systems. With regard to the ion implantation of the SESAM, we are grateful to Dr. Todd Simpson of the University of Western Ontario for his assistance. In addition, Mr. Shane Boisclair of the NRC Optical Components Laboratory, Mr. Ed Vilks and Mr. Martin Byloos of the Institute for Microstructural Sciences provided valuable technical contributions. The authors thank Dr. J.-S. Boulanger and Mr. S. Cundy for providing and monitoring the H-maser reference signal. This work was partially supported by the Canadian Institute for Photonics Innovations (CIPI).

References and Links

1. Eckstein JN, Ferguson AI, Hänsch TW. High-resolution two-photon spectroscopy with picosecond light pulses. Phys. Rev. Lett. 1978; 40:847–850.
2. Udem, Th.; Reichert, J.; Holzwarth, R.; Hänsch, TW. Absolute optical frequency measurement of the Cesium $D_1$ line with a mode-locked laser. Phys. Rev. Lett. 1999; 82:3568–3571.
3. Reichert J, Holzwarth R, Udem Th. Hänsch TW. Measuring the frequency of light with mode-locked lasers. Optics. Comm. 1999; 172:59–68.
4. Telle HR, Steinmeyer G, Dunlop AE, Stenger J, Sutter DH, Keller U. Carrier-envelope offset phase control: a novel concept for absolute optical frequency measurement and ultrashort pulse generation. Appl. Phys. B. 1999; 69:327–332.
5. Jones DJ, Diddams SA, Ranka JK, Stentz A, Windeler RS, Hall JL, Cundiff ST. Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis. Science. 2000; 288:635–639. [PubMed: 10784441]
6. Rauschenberger J, Fortier TM, Jones DJ, Ye J, Cundiff ST. Control of the frequency comb from a mode-locked Erbium-doped fiber laser. Optics Express. 2002; 10:1404–1410. http://www.opticsexpress.org/abstract.cfm?URI=OPEX-10-24-1404. [PubMed: 19452006]
7. Kim K, Washburn BR, Wipers G, Oates CW, Hollberg L, Newbury NR, Diddams SA. Stabilized frequency comb with a self-referenced femtosecond Cr:forsterite laser. Opt. Lett. 2005; 30:932–934. [PubMed: 15865403]
8. Czajkowski A, Madej AA, Dubé P. Development and study of a 1.5 $\mu$m optical frequency standard referenced to the P(16) saturated absorption line in the ($\nu_1+\nu_3$) overtone band of $^{13}\text{C}_2\text{H}_2$. Opt. Comm. 2004; 234:259–268.
9. Czajkowski A, Bernard JE, Madej AA, Windeler RS. Absolute frequency measurement of acetylene transitions in the region of 1540 nm. Appl. Phys. B. 2004; 79:45–50.
10. Quinn TJ. Practical realization of the definition of the metre, including recommended radiations of other optical frequency standards (2001). Metrologia. 2003; 40:103–133.
11. Keller U, Weingarten KJ, Kärtner FX, Kopf D, Braun B, Jung ID, Fluck R, Höninger C, Matuschek N. Aus der Au J. Semiconductor saturable absorber mirrors (SESAM’s) for femtosecond to nanosecond pulse generation in solid-state lasers. IEEE Journal of Selected Topics in Quantum Electron. 1996; 2:435–453.
12. Tsuda S, Knox WH, Cundiff ST, Jan WY, Cunningham JE. Mode-locking ultrafast solid-state lasers with saturable Bragg reflectors. IEEE Journal of Selected Topics in Quantum Electron. 1996; 2:454–464.
13. Naumov S, Sorokin E, Kalashnikov VL, Tempea G, Sorokina IT. Self-starting five optical cycle pulse generation in Cr$^{4+}$:YAG laser. Appl. Phys. B. 2003; 76:11–13.
14. Alcock J, Poole P, Sullivan BT. Hybrid semiconductor saturable absorber mirrors as passive mode-locking elements. Proc. SPIE. 2002; TD01:12–14.
15. Lederer MJ, Luther-Davies B, Tan HH, Jagadish C. GaAs based anti-resonant Fabry-Perot saturable absorber fabricated by metal organic vapor phase epitaxy and ion implantation. Appl. Phys. Lett. 1997; 70:3428–3430.
16. Onae, A.; Okumura, K.; Hong, F-L.; Matsumoto, H.; Nakagawa, K. Digest of the Conference on Precision Electromagnetic Measurements. IEEE Press; Piscataway NJ: 2004. Accurate frequency atlas of 1.5 μm band of acetylene measured by a mode-locked laser; p. 666-667.

17. Edwards CS, Margolis HS, Barwood GP, Lea SN, Gill P, Rowley WRC. High-accuracy frequency atlas of $^{13}$C$_2$H$_2$ in the 1.5 μm region. Appl. Phys. B. 2005; 80:977–983.

18. A. A. Madej, J.E. Bernard, A. J. Alcock, A. Czajkowski and S. Chepurov are preparing a manuscript to be called, “Accurate absolute frequencies of the $\nu_1+\nu_3$ band of $^{13}$C$_2$H$_2$ determined using an infrared mode-locked CnYAG laser frequency comb.”

Opt Express. Author manuscript; available in PMC 2010 October 19.
Fig. 1.
Schematic diagram of the cavity stabilized Cr$^{4+}$:YAG laser system. CM, chirped mirror; PZT, piezo-electric translator.
Fig. 2.
Autocorrelation traces and spectra for the Cr\textsuperscript{4+}:YAG laser; (a,b) Kerr lens mode-locked operation (c,d) SESAM stabilized mode-locking. The indicated FWHM pulse durations are based on a sech\textsuperscript{2} pulse shape. For comparison the absorption spectrum for C\textsubscript{2}H\textsubscript{2} is shown in (d).
Fig. 3.
Structure of the hybrid SESAM device, (a) intermediate structure (b) final structure.
Fig. 4.
Dynamic response of the SESAM before (a) and after (b) ion implantation.
Fig. 5. Schematic of the experimental setup used to measure the beats between the Cr:YAG comb and the radiation from two acetylene stabilized external cavity diode lasers.
Table 1

Summary of measured frequency intervals using the Cr:YAG frequency comb for selected $^{13}\text{C}_2\text{H}_2$ reference lines relative to a standard laser stabilized to the P(16) line. Also shown is the determined absolute frequency of each line. The results are compared with absolute Ti:Sapphire comb measurements (in parentheses) of the same reference lines as described in Ref. [9].

| Line | Line - P(16) [kHz] | Line Frequency [kHz] | Difference [kHz] |
|------|-------------------|----------------------|-----------------|
| P(10) | 452 257 032.7 ± 1.1 | 194 821 826 415.2 ± 1.7 | −1.4 ± 1.9 |
|       |                   | (194 821 826 416.66 ± 0.74) |                |
| P(11) | 378 572 272.5 ± 2.1 | 194 748 141 655.0 ± 2.5 | −0.1 ± 2.6 |
|       |                   | (194 748 141 655.15 ± 0.40) |                |
| P(15) | 77 063 007.0 ± 1.1  | 194 446 632 389.5 ± 1.7  | −1.8 ± 1.7 |
|       |                   | (194 446 632 391.28 ± 0.36) |                |
| P(20) | −314 976 289.9 ± 1.1 | 194 054 593 092.6 ± 1.7  | + 0.4 ± 1.7 |
|       |                   | (194 054 593 092.22 ± 0.38) |                |
| P(21) | −395 402 885.4 ± 1.1 | 193 974 166 497.1 ± 1.7  | −1.1 ± 1.9 |
|       |                   | (193 974 166 498.21 ± 0.80) |                |