Coherent heterodyne-assisted pulsed spectroscopy: sub-Doppler two-photon spectra of krypton, characterizing a tunable nonlinear-optical ultraviolet light source

Y. He · M. Kono · R.T. White · M.J. Sellars ·
K.G.H. Baldwin · B.J. Orr

Received: 5 January 2010 / Revised version: 1 March 2010 / Published online: 14 May 2010
© Springer-Verlag 2010

Abstract A chirp-minimized, nanosecond-pulsed system has been developed to generate tunable coherent ultraviolet light at ~ 212.5 nm by fourth-harmonic conversion of output from an amplified, injection-seeded optical parametric oscillator (OPO). Our CHAPS (coherent heterodyne-assisted pulsed spectroscopy) method is used to characterize the frequency stability and optical bandwidth of the system's output radiation by recording sub-Doppler two-photon excitation spectra of krypton. In our new scanned-reference variant of CHAPS, the central frequency for each amplified OPO pulse is logged by the optical-heterodyne chirp-analysis software, with the frequency of the seed laser slowly tuned and monitored by a high-resolution wavemeter, unlike our previous implementation of CHAPS where the seed-laser frequency was fixed. For the amplified, up-converted pulses at ~ 212.5 nm, our CHAPS measurements indicate an optical bandwidth of ~ 100 MHz, consistent with the Fourier-transform limit of their duration (~ 4.5 ns).

1 Introduction

We have previously developed a high-performance all-solid-state nonlinear-optical (NLO) light source to produce narrowband tunable pulses of coherent light in the range of 820–850 nm with ≤ 5 μ J energy, as a versatile source of tunable single-longitudinal-mode (SLM) coherent light for high-resolution spectroscopy [1–7]. It is useful in applications for which high peak power and narrow optical bandwidth are needed (e.g., in tens-of-ns pulses), especially in the vacuum ultraviolet (VUV) region where NLO up-conversion is essential.

For instance, our system will generate narrowband VUV light at ~ 120 nm (by frequency-quadrupling light at ~ 840 to ~ 210 nm, then two-photon-resonant four-wave mixing in krypton, Kr) to measure the $2^1S \leftarrow 1^1S$ two-photon transition of helium (He) as a test of quantum-electrodynamics theory. Previous studies, using a continuous-wave (cw) tunable Ti:sapphire laser and a pulsed dye amplifier [8–10], yielded limited bandwidth and uncertainties attributable to population-inversion and refractive-index variations in the dye amplifier. We expect our new NLO VUV light source to circumvent such problems.

We use a high-performance pulsed optical parametric oscillator (OPO) based on periodically poled KTiOPO₄ (PPKTP), pumped by a long-duration (~ 27 ns) pulsed SLM Nd:YAG laser (Continuum Powerlite 9000) and injection-seeded by a cw Ti:sapphire ring laser (Coherent 899-21) [5, 6], to generate tunable SLM coherent light in the wavelength range of 820–850 nm. Optical amplification and fourth-harmonic generation (4HG) then yield up-converted ultraviolet pulsed radiation. To validate the spectroscopic utility of this all-solid-state tunable SLM source of coherent pulsed ultraviolet radiation, we measure the sub-Doppler spectral profile of a two-photon resonance in Kr gas at ~ 212.55 nm.

Y. He · B.J. Orr (✉)
MQ Photonics Research Centre, Department of Physics and
Engineering, Macquarie University, Sydney, NSW 2109,
Australia
e-mail: b.orr@physics.mq.edu.au
Fax: +61-2-9850-8289

M. Kono · M.J. Sellars · K.G.H. Baldwin
Research School of Physics and Engineering, The Australian
National University, Canberra, ACT 0200, Australia

R.T. White
Institute for Photonics and Advanced Sensing (IPAS), University
of Adelaide, Adelaide, SA 5005, Australia

Fig. 1 Optical-heterodyne-detected chirp-analysis software [7] provides a real-time display of the recorded beat waveform (*top, left*) and its Fourier transform (with filter function; *top and center, right*), the reconstructed profile of the amplified OPO signal pulse (*center, left*), the calculated instantaneous-frequency profile (*bottom, left*), and cal-

culated pulse durations and chirp parameters (*bottom right*). In the example shown, the OPO is well seeded with frequency chirp <10 MHz (between 50% intensity points—table, lower right). It is remarkable that optical amplification does not cause the frequency chirp to deteriorate

As previously reported [3–7], the primary OPO signal output at ~ 850 nm has well-characterized shot-to-shot frequency stability and minimal frequency chirp, with its optical bandwidth approaching the sub-20-MHz Fourier-transform limit for a pulse duration of ~ 25 ns. An optical-heterodyne detection (OHD) technique [1–7] is able to monitor frequency chirp, fluctuations in the temporal profiles, and central-frequency shifts for successive optical pulses, thereby minimizing the spectroscopic limitations of such operating characteristics.

Sub-Doppler spectroscopic measurements play a key role in verifying the performance of this pulsed tunable NLO system at successive stages of its operation. For example, the fundamental OPO signal output has previously been characterized *via* sub-Doppler two-photon $8S \leftarrow 6S$ spectra of cesium (Cs) vapor at ~ 822.5 nm [5, 6]. To do this, we devised the CHAPS (Coherent Heterodyne-Assisted Pulsed Spectroscopy) approach, with the reference frequency of the cw Ti:sapphire seed laser held fixed on a Cs resonance peak and our OHD method used to log the central frequency of each OPO pulse (pre- or post-OPA) and record a sub-Doppler spectrum spanned by the OPO or OPA frequency jitter.

We report a novel scanned-reference variant of CHAPS, in which the cw seed-laser frequency is slowly tuned and monitored by a high-resolution wavemeter. By controlled tuning of the cw seed laser and logging the central frequency for each amplified OPO pulse at ~ 850.2 nm by means of our OHD real-time chirp-analysis software, we can correct any pulse-to-pulse OPO or amplifier frequency fluctuations and

produce sub-Doppler CHAPS spectra of the $4p^5 5p[1/2]_0 \leftarrow 4p^6$ two-photon resonance in Kr at ~ 212.55 nm [11–13], over a frequency range exceeding that spanned by frequency jitter of the pulsed OPO.

2 Experimental

To amplify the OPO output at ~ 850 nm prior to NLO up-conversion, we previously used a LiNbO₃ optical parametric amplifier (OPA). This OPA provided a pulse-energy gain of ~ 8 , but the optical pulse duration was reduced from ~ 23 to ~ 15 ns FWHM, degrading the Fourier-transform-limited optical bandwidth [5, 6]. To achieve much higher optical power ($\sim 8000 \times$ gain), we have installed an advanced two-stage Ti:sapphire amplifier (Amplitude Technologies), emulating recent work [14] on amplification of SLM Ti:sapphire laser output. The booster and multipass stages of this amplifier (pumped at 532 nm by ~ 35 -mJ and ~ 200 -mJ pulses from our long-pulse Nd:YAG laser) increase the SLM OPO signal output pulse energies from ≤ 5 μ J to ~ 1 mJ and ~ 40 mJ, respectively, with good spatial beam profile.

As in previous work [1–7], we measure OHD beats between amplified OPO pulses and cw light from the seed laser (modulator-shifted by ~ 730 MHz), to characterize frequency-chirp effects due to time-dependent optical phase variations during the OPO pulse and in subsequent amplification stages. We have previously shown [1–7] how to

Fig. 2 Temporal profiles of amplified fundamental and up-converted 4HG pulses (at ~ 850.2 nm and ~ 212.55 nm, respectively), measured by a fast photodetector (Alphas, model UPD-200-UD, 2.0-GHz bandwidth, <0.2 -ns rise-time)

minimize frequency chirp by operating the OPO system at its phase-matching wavelength (when the free-running OPO and seed wavelengths coincide). Comparable measures have been adopted to optimize instantaneous-frequency characteristics of the Ti:sapphire amplifier output, as depicted in Fig. 1, which is a representative frame generated by our OHD software. The chirp is remarkably small (well below 10 MHz FWHM) but the amplified pulse duration is disappointingly short (≤ 6 ns FWHM, compared with ~ 25 ns for the OPO output pulse itself). This four-fold optical-bandwidth deterioration seems to arise from extraction of most of the gain of the Ti:sapphire pulsed amplifier by the leading part of each OPO pulse; this problem is being addressed in pursuit of higher spectroscopic resolution.

The SLM pulsed output at ~ 850.2 nm from the Ti:sapphire-amplified OPO is up-converted by 4HG to ~ 212.55 nm in two second-harmonic-generation crystals: LiB_3O_5 , followed by $\beta\text{-BaB}_2\text{O}_4$. A pulse energy of ~ 40 mJ at ~ 850.2 nm yields ~ 1 mJ/pulse at ~ 212.55 nm. Temporal profiles of fundamental and 4HG pulses, measured by a 2.0-GHz photodetector, are shown in Fig. 2; their FWHM durations are ~ 6 and ~ 4.5 ns, respectively. The profile of the pulse at ~ 212.55 nm is quasi-Gaussian; its corresponding Fourier-transform-limited optical bandwidth would be ~ 100 MHz.

The pulsed coherent light at ~ 212.55 nm (~ 3 -mm diameter) propagates to the Kr ionization cell, where it is collimated and $3\times$ -expanded by a two-lens Galilean telescope. This parallel, unfocused beam is retro-reflected back through the cell by a flat mirror. With pure Kr gas in the cell, the two-photon-resonant ionization signal is collected by a longitudinal electrode with a DC bias voltage of 36 V and a

Fig. 3 Two-photon-resonant ionization spectra of the $4p^5 5p[1/2]_0 \leftarrow 4p^6$ transition in Kr gas, recorded by means of our chirp-minimized narrowband tunable source of coherent ultraviolet light at ~ 212.55 nm: (a) a wide-ranging scan of the spectral profile, comprising a sub-Doppler peak (~ 150 -MHz FWHM) superimposed upon a Doppler-broadened pedestal (~ 2 -THz FWHM); (b) a more detailed scan of the sub-Doppler peak with the boxcar-detected ionization signal plotted without correcting for frequency fluctuations in the amplified OPO pulse; (c) data corresponding to trace (b), following CHAPS analysis to determine the central frequency of each pulse relative to the (tuned) cw Ti:sapphire seed-laser frequency. The pulsed optical power is typically <0.1 MW cm^{-2} and Kr pressures are: (a) 0.13 Torr (17 Pa); (b, c) 0.10 Torr (13 Pa). Superimposed on traces (b) and (c) is a tentatively simulated spectrum, based on known Kr isotopic shifts and abundances [14, 15]

boxcar integrator to record survey spectra. The Kr pressure and incident optical power (varied by a polarizing attenuator) were carefully optimized to record reliable, saturation-free two-photon-resonant ionization signals.

3 Results and discussion

Figure 3 shows spectra obtained for the $4p^5 5p[1/2]_0 \leftarrow 4p^6$ two-photon-resonant ionization process in Kr gas, as a function of the tuned 4HG frequency (four times the frequency of the amplified OPO signal output). The broad spectral profile in Fig. 3(a) shows the Doppler-broadened pedestal on which the sub-Doppler peak is superimposed. Figures 3(b) and 3(c) are more detailed scans of the sub-Doppler peak, without

and with CHAPS-type corrections for frequency fluctuations in the amplified OPO pulse, respectively.

The synthetic spectrum, superimposed in Figs. 3(b) and 3(c), takes account of Kr isotopic components that are separated by intervals of 50–80 MHz on the 4HG frequency scale [14, 15], with relative intensities that reflect the natural abundance of the major isotopes of Kr (57.0% ^{84}Kr , 17.3% ^{86}Kr , 11.6% ^{82}Kr , 11.5% ^{83}Kr) but do not allow for possible isotopic variation in the two-photon transition probability. The simulation allows crudely for the spectral bandwidth (~ 100 MHz) derived from the pulse duration (~ 4.5 -ns) and the underlying Doppler pedestal from Fig. 3(a), but neglects pressure broadening (≤ 5 MHz FWHM [16]).

The ~ 100 -MHz FWHM linewidth of the coherent light at ~ 212.55 nm is limited predominantly by the ~ 4.5 -ns optical pulse duration and does not allow clear resolution of the Kr isotopic structure in the $4p^5 5p[1/2]_0 \leftarrow 4p^6$ two-photon excitation spectrum. We note that these spectra were recorded with minimal frequency chirp (≤ 10 MHz at ~ 850 nm) and with small shot-to-shot fluctuations (~ 4 -MHz standard deviation at ~ 850 nm) in the central frequency. The fit of the simulated curve to the CHAPS-analyzed data in Fig. 3(c) is therefore not markedly superior to that for the boxcar-detected data in Fig. 3(b).

4 Conclusion

We have measured two-photon-resonant ionization spectra of Kr at ~ 212.55 nm to characterize a nearly Fourier-transform-limited OPO light source following amplification and 4HG frequency conversion, using a new scanned-reference variant of CHAPS. The spectra obtained are consistent with the optical bandwidth and shot-to-shot frequency stability determined by optical-heterodyne measurements, which indicate that the linewidth is dominated by the relatively short pulse duration (~ 4.5 ns FWHM). In ongoing research, we intend to circumvent the Ti:sapphire amplifier system's severe pulse-shortening effect and attain significantly higher sub-Doppler spectroscopic resolution.

Our progress towards measurement of the fundamentally important $2^1\text{S} \leftarrow 1^1\text{S}$ two-photon spectrum of He by generating ~ 120 -nm tunable SLM coherent light (enhanced by the same two-photon resonance in Kr) complements recent advances elsewhere [13, 14, 17, 18], aiming to address

discrepancies between state-of-the-art theory [19, 20] and earlier experimental estimates [8, 21] of the Lamb shift in ground-state He.

Acknowledgements We acknowledge funding from the Australian Research Council and thank Drs S. Cavanagh and W. Krolikowski for experimental assistance. The Ti:sapphire amplifier system used was custom-built for us by Amplitude Technologies (France).

References

1. R.T. White, Y. He, B.J. Orr, M. Kono, K.G.H. Baldwin, *Opt. Lett.* **28**, 1248 (2003)
2. R.T. White, Y. He, B.J. Orr, M. Kono, K.G.H. Baldwin, *J. Opt. Soc. Am. B* **21**, 1577 (2004)
3. R.T. White, Y. He, B.J. Orr, M. Kono, K.G.H. Baldwin, *J. Opt. Soc. Am. B* **21**, 1586 (2004)
4. R.T. White, Y. He, B.J. Orr, M. Kono, K.G.H. Baldwin, *Opt. Express* **12**, 5655 (2004)
5. M. Kono, K.G.H. Baldwin, Y. He, R.T. White, B.J. Orr, *Opt. Lett.* **30**, 3413 (2005)
6. M. Kono, K.G.H. Baldwin, Y. He, R.T. White, B.J. Orr, *J. Opt. Soc. Am. B* **23**, 1181 (2006)
7. R.T. White, Y. He, B.J. Orr, M. Kono, K.G.H. Baldwin, *J. Opt. Soc. Am. B* **24**, 2601 (2007)
8. S.D. Bergeson, A. Balakrishnan, K.G.H. Baldwin, T.B. Lucatorto, J.P. Marangos, T.J. McIlrath, T.R. O'Brian, S.L. Rolston, C.J. Sansonetti, J. Wen, N. Westbrook, C.H. Cheng, E.E. Eyler, *Phys. Rev. Lett.* **80**, 3475 (1998)
9. S.D. Bergeson, A. Balakrishnan, K.G.H. Baldwin, T.B. Lucatorto, J.P. Marangos, T.J. McIlrath, T.R. O'Brian, S.L. Rolston, C.J. Sansonetti, J. Wen, N. Westbrook, C.H. Cheng, E.E. Eyler, *Phys. Scr.*, **T 83**, 76 (1999)
10. S.D. Bergeson, K.G.H. Baldwin, T.B. Lucatorto, T.J. McIlrath, C.H. Cheng, E.E. Eyler, *J. Opt. Soc. Am. B* **17**, 1599 (2000)
11. J.P. Marangos, N. Shen, H. Ma, M.H.R. Hutchinson, J.P. Connerade, *J. Opt. Soc. Am. B* **307**, 400 (2005)
12. F. Brandi, W. Hogervorst, W. Ubachs, *J. Phys. B: At. Mol. Opt. Phys.* **35**, 1071 (2002)
13. S. Witte, R.T. Zinkstok, W. Ubachs, W. Hogervorst, K.S.E. Eikema, *Science* **307**, 400 (2005)
14. S. Hannemann, E.-J. van Duijn, W. Ubachs, *Rev. Sci. Instrum.* **78**, 103102 (2007)
15. F. Brandi, W. Hogervorst, W. Ubachs, *J. Phys. B: At. Mol. Opt. Phys.* **35**, 1071 (2002)
16. D.A. Jackson, *J. Opt. Soc. Am.* **71**, 1064 (1981)
17. E.E. Eyler, D.E. Chieda, M.C. Stowe, M.J. Thorpe, T.R. Schibli, J. Ye, *Eur. Phys. J. D* **48**, 43 (2008)
18. K.S.E. Eikema, Private communication, Vrije Universiteit, Amsterdam, 2009
19. K. Pachucki, *Phys. Rev. A* **74**, 022512 (2006)
20. D.C. Morton, Q. Wu, G.W.F. Drake, *Canad. J. Phys.* **84**, 83 (2006)
21. K.S.E. Eikema, W. Ubachs, W. Vassen, W. Hogervorst, *Phys. Rev. A* **5**, 1866 (1997)