



<http://www.diva-portal.org>

Postprint

This is the accepted version of a paper published in *Laser Physics Letters*. This paper has been peer-reviewed but does not include the final publisher proof-corrections or journal pagination.

Citation for the original published paper (version of record):

Persson, A., Berglund, M., Thornell, G., Possnert, G., Salehpour, M. (2014)

Stripline split-ring resonator with integrated optogalvanic sample cell.

Laser Physics Letters, 11(4): 045701

<http://dx.doi.org/10.1088/1612-2011/11/4/045701>

Access to the published version may require subscription.

N.B. When citing this work, cite the original published paper.

Permanent link to this version:

<http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-218769>

Stripline split-ring resonator with integrated optogalvanic sample cell

Anders Persson^{1,3}, Martin Berglund², Greger Thornell², Göran Possnert¹ and Mehran Salehpour¹

¹Department of Physics and Astronomy, Ion Physics, Uppsala University, Uppsala, SE-751 20, Sweden.

²Department of Engineering Sciences, Ångström Space Technology Centre, Uppsala University, Uppsala, SE-751 21, Sweden.

³Corresponding author.

This document is the Post-print version of a Published Work that appeared in final form in *Laser Physics Letters*. It should be cited as “Anders Persson *et al* 2014 *Laser Phys. Lett.* **11** 045701 doi:10.1088/1612-2011/11/4/045701”. To access the final edited and published work see: <http://iopscience.iop.org/1612-202X/11/4/045701/>

E-mail: anders.persson@physics.uu.se

Short title: Stripline split-ring resonator with integrated optogalvanic sample cell

Abstract. Intracavity optogalvanic spectroscopy (ICOGS) has been proposed as a method for unambiguous detection of rare isotopes. Of particular interest is ^{14}C , where detection of extremely low concentrations in the $1:10^{15}$ range ($^{14}\text{C}:^{12}\text{C}$), is of interest in, e.g., radiocarbon dating and pharmaceutical sciences. However, recent reports show that ICOGS suffers from substantial problems with reproducibility. To qualify ICOGS as an analytical method, more stable and reliable plasma generation and signal detection are needed. In our proposed setup, critical parameters have been improved. We have utilized a stripline split-ring resonator microwave-induced microplasma source to excite and sustain the plasma. Such a microplasma source offers several advantages over conventional ICOGS plasma sources. For example, the stripline split-ring resonator concept employs separated plasma generation and signal detection, which enables sensitive detection at stable plasma conditions. The concept also permits *in situ* observation of the discharge conditions, which was found to improve reproducibility. Unique to the stripline split-ring resonator microplasma source of in this study, is that the optogalvanic sample cell has been embedded in the device itself. This integration enabled improved temperature control and more stable and accurate signal detection. Significant improvements are demonstrated, including reproducibility, signal-to-noise ratio and precision.

PACS: 52.38.Dx, 52.50.Dg, 52.70.Ds, 52.80.Pi, 52.80.Yr, 42.55.Lt, 42.62.Fi, 42.62.Be.

Keywords: Optogalvanic spectroscopy, Laser-assisted ratio analyzer, Split-ring resonator, microwave-induced microplasma source

1. Introduction

Laser spectroscopy utilizing the optogalvanic (OG) effect has long been a powerful method for the investigation of atomic and molecular species [1]. In the 1990^{ths}, the applicability OG spectroscopy was extended to cover the isotopic composition of molecules, more precisely the $^{13}\text{C}/^{12}\text{C}$ ratio in CO_2 [2], using the so called laser-assisted ratio analyzer (LARA) technique. With the advent of intracavity optogalvanic spectroscopy (ICOGS) [3, 4], even measurements of $^{14}\text{C}/^{12}\text{C}$ ratios were suggested to be feasible. With a claimed detection limit in the 10^{-15} range, ICOGS could be applied to, e.g., radiocarbon dating in archaeology [5], measurements of cell turnover in biomedicine [6], and microdosing in drug development [7]. However, we have recently shown that ICOGS suffers from considerable problems with the stability and reproducibility of discharge conditions in the internal plasma cell [8, 9], making the method inadequate quantitative measurements.

In another report, we recently demonstrated the first ever use of a split-ring resonator microwave-induced microplasma source (MPS) for OG spectroscopy [10]. More precisely, the MPS was a stripline split-ring resonator (SSRR), in which a microwave plasma is generated between the ends of a stripline dipole, folded into a circular shape [11].

SSRRs offer several advantages over MPSs in general, and over plasma sources for OG spectroscopy in particular. For example, they offer a dense, yet cold plasma, which is advantageous for laser spectroscopy in the mid- and far-IR regimes [1]. Moreover, the plasma is generated between the ends of the dipole, which are located in the middle of a hole going through the device. The plasma forms an ionized sheet with a well controlled spatial distribution across hole. This reduces plasma erosion, otherwise causing chemical contamination of a spectral measurement, and extends the lifetime of the device. Even more importantly, the stripline concept offers improved resistance to electromagnetic interference compared with, e.g., split-ring resonators based on a microstrip design, thanks to the electromagnetic shielding by the two ground planes sandwiching the dipole. [11]

One of the intended applications of the SSRR is ICOGS. In ICOGS, the sample, in the form of CO_2 , is partly ionized in a plasma cell, also called a sample cell, and subjected to resonant radiation from a CO_2 laser beam. The molecules in the cell interact with the laser beam by either absorption or stimulated emission. This affects the impedance of the plasma, which is the measured entity in OG spectroscopy.

Conventional ICOGS measures the impedance change via a change in the energy going into the discharge, by monitoring the feedback circuit in the power amplifier driving the plasma [3, 4, 8, 9]. This method is almost 30 years old [12], and suffers from electromagnetic interference issues [9]. Moreover, the integration of the signal detection with the plasma power supply causes noise from the amplifier to be picked up in the signal detector, and makes the detector sensitive to, e.g., thermal fluctuations in the power supply. Hence, more stable and reliable plasma generation and signal detection are essential for further development of ICOGS [9].

We previously showed that the OG signal in an SSRR could be measured separately from the power supply, by inserting two probes in the plasma [10]. Separating the plasma generation from the signal detection, enables sensitive OG measurements under stable plasma conditions, and, hence, more reliable OG spectroscopy. However, this new measurements scheme still required improved stability and signal-to-noise ratio (SNR) to become competitive. In this paper we present the implementation of some significant improvements to the setup. Among those were replacing the laser with a single longitudinal mode CO_2 laser with an actively stabilized cavity, improving the electromagnetic

shielding and the cooling of the power supply, performing measurements in batch mode, i.e., at constant pressure, and, most importantly, embedding the sample cell inside the SSRR. The implications of these improvements on the reproducibility, stability, accuracy and SNR are carefully presented.

2. Materials and Methods

The design of the SSRR used in this study was identical to the SSRR with a 2 mm wide gap used in reference 10, except for two key features. Firstly, the hole through the gap of the SSRR was sealed off by two 1 mm thick, antireflection-coated Ge windows (Edmund Optics Ltd, York, UK), figure 1, thus creating a miniature sample cell inside the SSRR. This cell is referred to as an integrated SSRR cell. Secondly, two 80 mm long (1/4 in. diameter), Ni plated, stainless steel tubes (Swagelok, Cleveland, OH, USA) were connected to the fluidic channels of the SSRR by soldering, figure 1. The ends of both tubes were connected to valves, one of which was connected to a gas handling system (see figure 2). The other valve was kept closed throughout the experiments, but could be used to perform OG measurements on a sample flow. The total volume between the valves was 7.5 ml (Appendix A).

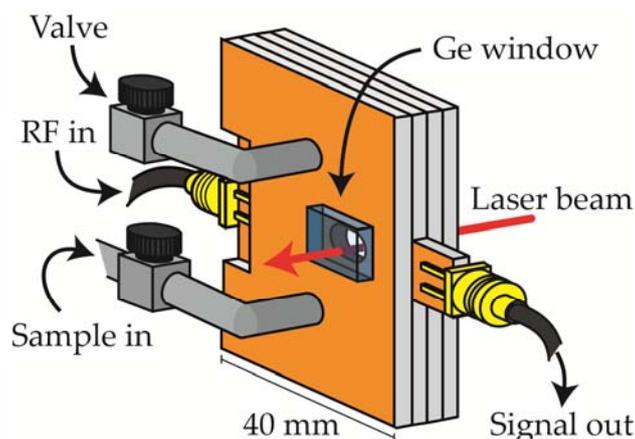


Figure 1. Drawing of an integrated SSRR cell with fluidic connections and laser transparent Ge windows.

The electronics used for generating the plasma also closely resembled that of reference 10. However, some substantial improvements were made. The data acquisition (DAQ) and the circulator, which were identified as sources of instabilities and noise in the RF wave feeding the plasma, were replaced by more stable components: a PCI 6120 with a BNC-2110 connector block (National Instruments Corp., Austin, TX, USA), and a CS 3.000 (e-MECA, Denville NJ, USA). Moreover, all the electronics, except for the DAQ and the computer, were mounted in an electromagnetically shielded, fan-cooled box. In addition to the forced convective cooling of the box interior, the main amplifier was attached to a heat sink with an additional fan for increased thermal stability.

The optics and gas handling of the system, figure 2, closely resembled those of reference 9, with some exceptions. For example, the $^{14}\text{CO}_2$ laser was removed, and the beams from the $^{12}\text{CO}_2$ and $^{13}\text{CO}_2$ lasers were redirected to go through/to the reference cell [8, 9], spectrum analyzer (CO₂ Laser Spectrum Analyzer, Macken Instruments, Santa Rosa, CA, USA), and thermopile power meter (Vega, Ophir Optronics Solutions, Jerusalem, Israel). The lasers – a $^{12}\text{CO}_2$ Merit-SL and a $^{13}\text{CO}_2$ L20-SZ (Access Laser Co., Everett, WA, USA) – were both single longitudinal mode lasers with active temperature

control, and piezoelectric tuning of the lasing frequency. A ZnSe lens, with a focal length of 150 mm (LA7028-F, Thorlabs Inc., Newton, NJ, USA), and a steerable mirror were added to the optics to properly align the laser beams through the cell. The gas handling system was identical to the one in reference 9, and could supply the cell with CO₂, N₂, or a mix of the two, at pressures ranging between 1 Pa (system base pressure) and 2500 Pa (upper limit of the pressure gauge). The total volume of the system was 62 ml excluding the SSRR (Appendix A).

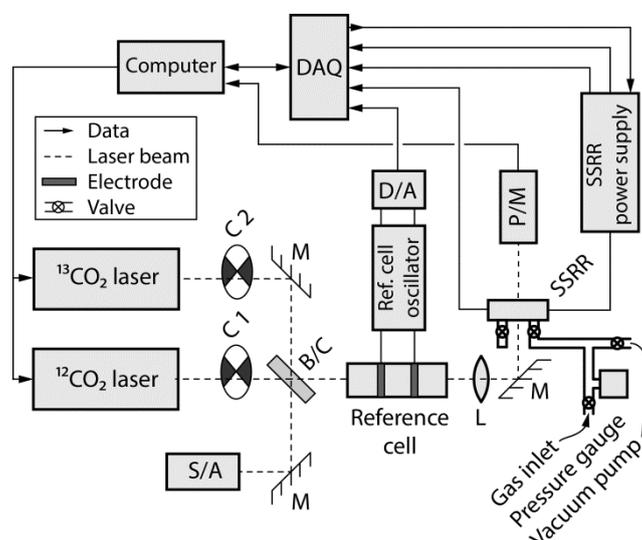


Figure 2. Schematics of the OG spectroscopy setup including optics, gas handling and electronics. Here, B/C denotes beam combiner, C chopper, D/A differential amplifier, DAQ data acquisition, L lens, M, mirror, P/M power meter, and S/A spectrum analyzer.

The DAQ had 4 analog inputs which were used to measure the OG signals from the SSRR and the reference cell, together with the power transmitted to and reflected from the SSRR. The signal from the reference cell was amplified by 40 dB in a differential amplifier before acquisition, whereas the signal from the SSRR was measured without amplification. Moreover, the analog and digital outputs of the DAQ were used to control the frequency and attenuation of the RF wave transmitted to the SSRR, whereas the piezoelectric tuning of the CO₂ lasers was controlled directly by the computer via an RS-232 interface. Similarly, the signal from the power meter was measured via an USB interface. All data handling was controlled by custom-made software (LabVIEW 2010 10.0, National Instruments Inc., Austin, TX, USA). The optics, gas handling, and electronics, together with the software, measurement procedure, and parameters of the different experiments, are more thoroughly described in Appendix A.

3. Results and discussion

The wavelength of the lasers could either be scanned over several laser transitions, by tuning the length of the laser cavity with the integrated piezoelectric control, or be locked to the peak of a particular laser transition (Appendix A). The intensity and wavelength of the laser beams were continuously monitored by the power meter and the spectrum analyzer. Figure 3 shows the OG signal from the SSRR, when the ¹³CO₂ laser was scanned through the upper part of the ¹²C¹⁶O₂ P band. Here, the OG signal has been scaled by the laser power (shown for comparison), to reveal the spectral distribution of the resonant absorption. A total of seven absorption peaks, corresponding to the transitions P36-P46 and P50, could be clearly distinguished when the cell was filled with either pure CO₂ or CO₂ mixed in N₂. However, when the cell was filled with pure N₂, no resonant absorption could be seen, only non-resonant background. This is more or less exactly what can be expected from

an OG spectrum at these conditions [8, 13]. Moreover, the OG signal from the SSRR correlated almost perfectly with that from the reference cell – with a coefficient of determination of up to $R^2 = 0.96$, regarding the reference cell data as a fit – wherefore the integrated SSRR cell was assumed to measure the OG response of the plasma in a reliable manner.

In reference 10, it was observed that direct heating of the SSRR from the laser beam could cause false OG signals, especially at low chopper frequency. Here, however, no such effects were seen, probably because of improved (convective) cooling of the device (no longer being contained in a vacuum chamber), and better alignment of the laser beams through the gap.

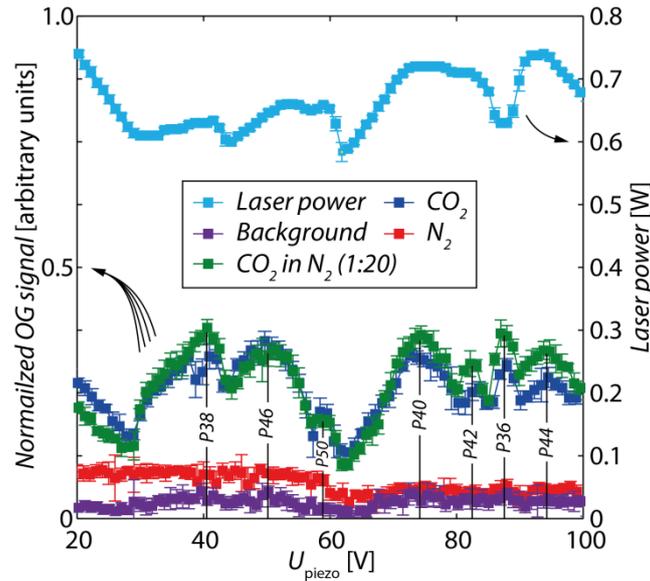


Figure 3. Spectra of different gases and mixtures acquired by scanning the wavelength of the $^{12}\text{CO}_2$ laser (tuning voltage). The measured OG signals were normalized by the laser power, which is shown for comparison.

In reference 10, turbulence and slipping in the flow through the SSRR were identified as a major source of instability and noise in the measurements. To avoid such instabilities, all measurements in this study were made at, virtually, zero flow. Since the ignition and maintenance of the plasma caused dissociation of molecules [8], and heating of the sample gas, the pressure, however, increased from its initial level until reaching a new equilibrium. To study the time to equilibrium, the pressure in the system was monitored with and without plasma. The inset in figure 4 shows the pressure change in CO_2 of nominally 5.0 mbar, where the change in absence of plasma corresponded to the system leak rate of 60 mPa/min. In the presence of plasma, the change reached a level corresponding to the leak rate, i.e. equilibrium, after about 45 min.

It can be assumed that a smaller volume, such as the integrated SSRR cell, will reach equilibrium faster. However, this could not be confirmed by measuring the pressure, since no pressure gauge was integrated in the cell. Instead, the power reflected from the plasma, P_R , which is very sensitive to variations in the plasma conditions, e.g., pressure variations, was used to monitor the time to equilibrium for both the closed and open cell, figure 4. When the whole system was filled to 5.0 mbar of CO_2 , the change in P_R too stabilized after about 45 min, confirming the assumption that P_R can be used to monitor equilibrium. However, on approaching equilibrium, P_R changed rapidly and intermittently, as seen from the peaks in the figure. If the SSRR cell was closed, the time to equilibrium was halved. Moreover, employing a mixture of CO_2 in N_2 (1:20) helped to further reduce

the time to equilibrium. At similar initial pressure, these times were about 25 min for the open, and 5 min for the closed cell, respectively. In this context, it should also be pointed out that the steel tubes constituted most of the volume in the integrated SSRR cell, and that, if valves could be integrated in the device itself, the time to equilibrium can be expected to be even shorter.

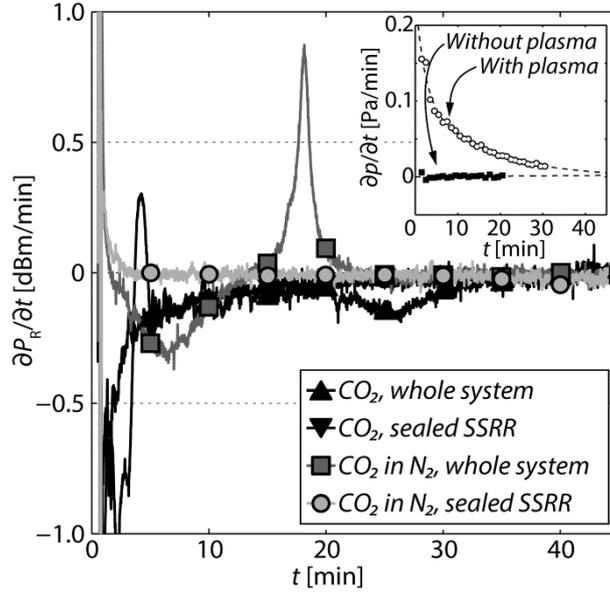


Figure 4. Reflected power variations after plasma ignition for different gas mixtures, where the integrated SSRR cell was either open or close. The inset shows the pressure change after ignition of a 5 mbar CO_2 plasma (white circles), and the leak rate of the system at similar conditions (black squares).

A common way to describe the stability of a measurement system is the so called Allan variance, σ_A^2 which relates to the time scale, τ , on which a signal can be regarded as constant, and, thus, the maximum time over which averaging should be performed. At small τ , σ_A^2 exhibits a τ^{-1} dependence, until, e.g., random-walk noise renders further averaging useless, and σ_A^2 starts tending to τ^1 . The optimum averaging time corresponds to the τ where σ_A^2 reaches its minimum. Figure 5 shows typical σ_A^2 for measurements at 5.0 mbar on CO_2 in N_2 (1:20) after different times of stabilization, with the SSRR cell closed. In these measurements the sampling rate was 1 Hz, wherefore τ can be translated directly into number of averages. As can be seen, the optimum number of averages depended on the time to equilibrium, discussed above, and reached up to 50 after the plasma had stabilized for >5 min. This is considerably better than what was reported in Refs. 9 and 10, probably due to the removal of flow instabilities compared with the former, and the more stable plasma generation and better signal detection schemes compared with the latter.

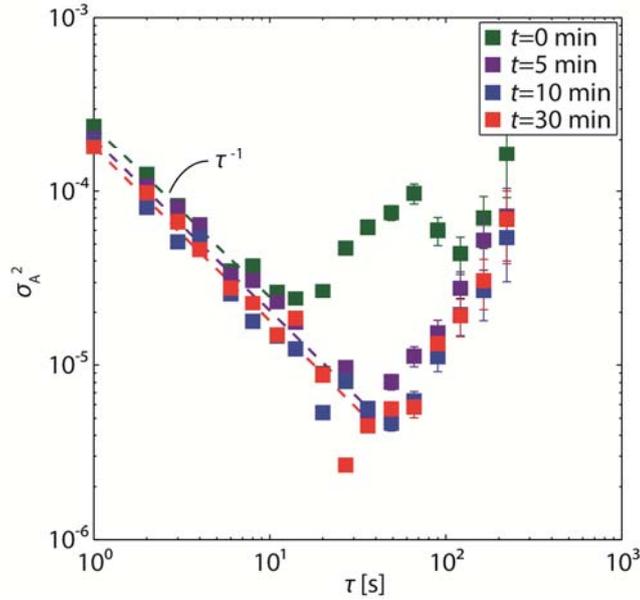


Figure 5. Allan variance, σ_A^2 , at different stabilization times, t , after plasma ignition for a 5 mbar plasma of CO₂ in N₂ (1:20). An optimum integration time, τ , corresponding to ~50 averages was reached after ~5 min stabilization.

The improved stability could also be seen in the SNR, as calculated from a Fourier transform of the OG signal from the closed SSRR (see of figure 6). This spectrum corresponds to a SNR of more than 40 dB, which is about 10 dB better than what was accomplished in reference 10. It should be pointed out that this SNR was achieved without amplification of the signal.

In reference 9, the irreproducibility of the discharge between consecutive measurements with identical initial conditions was identified as the principal limitation of ICOGS. In repeated experiments with identical initial pressure, stabilization time, plasma power, etc., the average level of the OG signal could differ by up to $\pm 30\%$ [9]. This uncertainty made quantitative analysis more or less impossible. In this context, an integrated SSRR cell offers two key advantages compared with conventional ICOGS sample cells. Firstly, the plasma generation and signal detection are separated. Hence, it is possible to control the stability of the plasma and the OG signal separately. This is impossible in an ordinary sample cell, where a stable plasma renders a weak signal and vice versa. Secondly, it is possible to monitor the discharge conditions in the SSRR cell in situ, by studying the power transmitted to and reflected from the plasma, and the voltage over the plasma probes.

Figure 6 shows the OG signal from the SSRR, normalized by the OG signal from the reference cell, in five consecutive measurements with identical initial conditions, after employing the same post-processing protocol as described in reference 9. Between each measurement, the plasma was turned off, and the gas handling system was evacuated. The OG signal is plotted versus P_R , which was used to determine when the plasma had stabilized. As can be seen, the reproducibility was greatly enhanced, with the average level of the different measurements differing by only around $\pm 3\%$ from the overall average ($\sigma = 0.029$). This is one order of magnitude better than what was reported in reference 9.

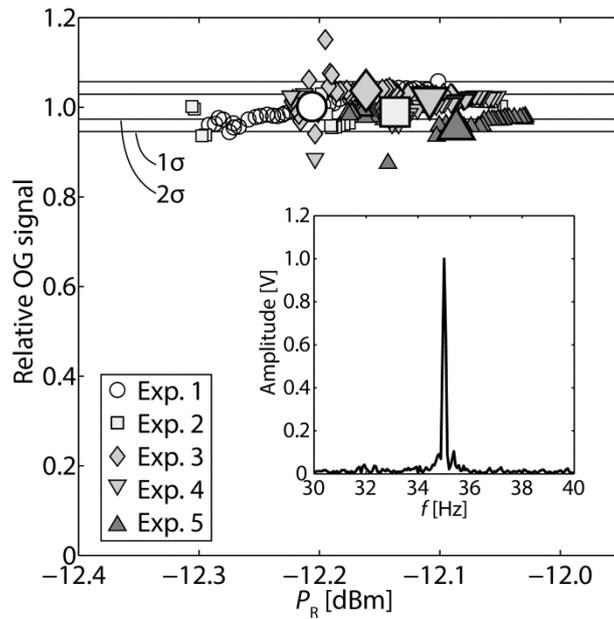


Figure 6. Reproducibility in five consecutive experiments with identical initial conditions (closed cell with 5.0 mbar CO₂ in N₂ mixed 1:20 plasma, stabilized for >5 min). The larger symbols show the average of each experiment, and the lines show 1 σ and 2 σ for the combined measurements. The inset shows a Fourier transform of the OG signal from the closed SSRR.

4. Conclusion

In conclusion, the applicability of an integrated SSRR cell for OG spectroscopy has been investigated. The cell was shown to double the time over which the signal was stable and averaging could be performed, and increased the SNR of the system by 10 dB, compared with previous work [10]. Moreover, the cell improved the reproducibility of the signal by one order of magnitude compared with traditional ICOGS plasma cells [9]. In addition to these substantial improvements, the SSRR concept provided enhanced control, by the separated plasma generation and signal detection, and *in situ* monitoring of the discharge conditions. Hence, the integrated SSRR cell will become a powerful tool in the investigation of ICOGS applicability to unambiguous ¹⁴C detection [3, 9]. However, even though the integrated SSRR cell offers substantial advantages for OG spectroscopy, it is still not fully optimized. Additional investigations into how the improved control of the plasma conditions can be utilized to optimize the stability and accuracy of the OG signal are required. Furthermore, different amplifying schemes should be investigated, together with methods for voltage or current biased probe measurements of the OG signal [11]. These studies are expected to improve the performance of the SSRRs even further.

Acknowledgements

The authors wish to acknowledge the Swedish Research Council for supporting this project (Reference A0442201).

Appendix A

Appendix A contains more thorough information on the hardware, software, measurement procedures, and experiments. In addition, the estimations of the internal volume of the integrated SSRR cell, and the gas handling system are justified.

A.1. Hardware

A schematic view of the hardware used in the presented studies is shown in Figure A1, with a close-up of the SSRR power supply.

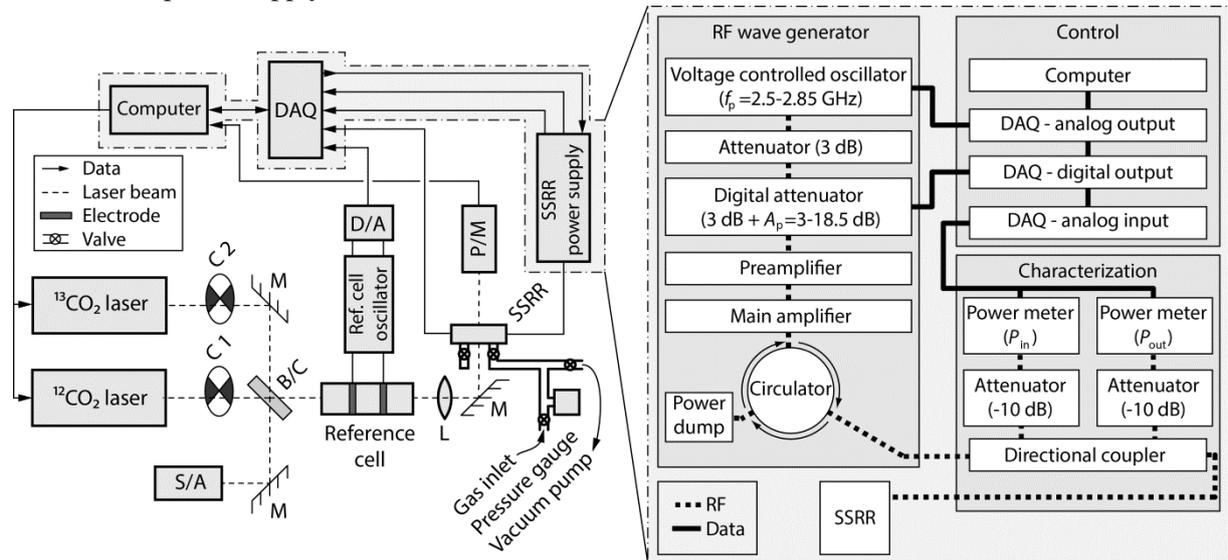


Figure A1. Schematic view of the hardware, including optics, gas handling and electronics (left), with a close-up of the SSRR power supply (right). Here, B/C denote beam combiner, C chopper, D/A differential amplifier, DAQ data acquisition, L lens, M, mirror, P/M power meter, and S/A spectrum analyzer.

A2. Software

The software used in this study was made in LabVIEW 2010 10.0 (National Instruments Inc., Austin, TX, USA). Four different programs were used to control the frequency and attenuation of the RF wave from the SSRR power supply, and the piezoelectric tuning of the lasers, as well as to measure the OG signal from the SSRR and reference cells along with the power transmitted to and reflected from the SSRR, and the laser power.

Program 1. The first program was used to measure the resonance frequency of the SSRR. This was done by measuring the transmitted and reflected power while sweeping the frequency of the RF wave from 2 to 3 GHz. The resonance frequency was determined by the global minimum of the reflected power. In this program, the attenuation was kept at the maximum 15.5 dBm, to avoid plasma ignition, which would disturb the measurement.

Program 2. The second program was used to ignite and maintain the plasma. The inputs to the program were attenuation and frequency, where the latter was the resonance frequency measured with Program 1. Both the attenuation and the frequency could be altered when the program was running, i.e., while the plasma was on. The plasma was ignited by 1.) setting the frequency to the resonance frequency, 2.) setting the attenuation to 0 dBm, 3.) turning on the voltage controlled oscillator (VCO). With the VCO on, RF was transmitted to the SSRR and the plasma was ignited. To avoid excess

dissociation and heating of the sample gas, the attenuation was changed back to 15.5 dBm shortly after ignition, at which point the plasma continued to burn.

Program 3. The third program was used to measure the OG signals while the wavelengths of the CO₂ lasers were scanned, using the piezoelectric tuning of the length of the laser cavity. This program was run in parallel with Program 2, and could be used to monitor if the plasma was on, by studying the voltage over the plasma probes in the SSRR.

The OG signals from the SSRR and the reference cell, along with the reflected and transmitted power were sampled with the DAQ, at a rate of 1 kHz. The sampled time series were then processed in different ways. In the case of the transmitted and reflected power, the averages of the series were recorded. In the case of the OG signals, the time series were transformed into the frequency domain using the Fourier transform. However, the OG signal from the SSRR had to be AC coupled before transformation. Moreover, the sampled time series were zero padded, to improve the resolution of the spectrum, and band-pass filtered between 10 and 90 Hz. The amplitude of the OG signal was finally determined by fitting a Gaussian distribution to a 6 Hz spectrum centered round the chopper frequency. The program had several input variables including sampling time, i.e., the time over which samples were collected, and chopper frequencies, i.e, the modulation rates of the laser beams. The laser power meter was sampled via the USB interface, at a rate of (sampling time)⁻¹.

The piezoelectric tuning was scanned in steps of 1 V, from 0 to 100 V, i.e., 101 steps. A third input to the program was the number of measurements that were recorded at each step. In total, Program 3 recorded the OG signals from the SSRR and reference cell at the ¹²CO₂ and ¹³CO₂ chopper frequencies, the transmitted and reflected power, the piezo-voltage, and the laser power, i.e., 8 outputs.

Program 4. The fourth program was used to measure the OG signals, while the laser wavelengths were kept constant, at a certain laser transition. This transition could be found either by manually tuning the piezoelectric control, or by using Program 3, with feedback from the OG signals and the spectrum analyzer. When centered at a transition, the piezo voltage was left constant. To avoid instabilities due to drift in the wavelength, the laser power was continuously monitored. However, the wavelength typically remained stable over time scales up to 1 h. The sampling was then performed in the same way as in Program 3, except for changing the piezo voltage, meaning that Program 4 only had 7 outputs.

A3. Measurement procedure

A measurement started by evacuating the gas handling system and cell to the base pressure. The sample gas was then introduced into the system to an adequate pressure. In the case of mixtures of CO₂ and N₂, a mixing ratio of 1:20 was employed. To achieve this, the system was firstly filled with 1 mbar of CO₂, followed by 19 mbar of N₂. The mixture was then allowed to stabilize for ~2 min, after which the system was pumped to the desired initial pressure. When the initial pressure was set, the plasma was ignited using Program 2. The plasma was then allowed to stabilize (unless the stability was the subject of the study), before a measurement was started using Programs 3 or 4.

A4. Experiments

The details of the experiments of the presented study are given in table A2. Here, the experiments are ordered after the figures in which they are presented. In the case of more than one experiment being presented in a single figure, several entries have been made.

Table A2. Details of the experiments ordered after the figure in which they are presented.

Experiment	Software	Gas	Pressure [mbar]	Sampling time [s]	Number of data points	Chopper frequency [Hz]	Laser transition
Figure 3	Program 3	CO ₂	5.0	1	8 ^a	77	N/A
	Program 3	N ₂	5.0	1	8 ^a	77	N/A
	Program 3	CO ₂ in N ₂ (1:20)	5.0	1	8 ^a	77	N/A
Figure 4	Program 4	CO ₂	5.0	1	2700	N/A	N/A
	Program 4	CO ₂ in N ₂ (1:20)	5.0	1	2700	N/A	N/A
Figure 4, inset	Program 4	CO ₂	5.0	60	20/30	N/A	N/A
Figure 5	Program 4 ^b	CO ₂ in N ₂ (1:20)	5.0	2	1000	77	P40
Figure 6	Program 4	CO ₂ in N ₂ (1:20)	5.0	2	50	35	P40
Figure 6, inset	Program 4	CO ₂ in N ₂ (1:20)	5.0	10	1	35	P40

^aNumber of measurements at each step of the piezoelectric control.

^bAllan variances were calculated using the Matlab script Allan.m

(<http://www.mathworks.se/matlabcentral/fileexchange/26659-allan-v3-0/content/allan.m>)

A5. Volume estimation

To estimate the internal volume of the gas handling system and the integrated SSRR cell, CO₂ gas of a known pressure (10.0 mbar) from a vessel with a known volume (2 ml) was introduced into the evacuated system with the cell open and closed. With the cell opened, the pressure in the whole system became 0.280 mbar, and with the cell closed the pressure became 0.313 mbar. Assuming constant temperature, and disregarding any leaks, the corresponding volumes could be calculated using the ideal gas law. In this way, the volume of the gas handling system was estimated to 62 ml, and the volume of the integrated SSRR cell including piping to 7.5 ml.

References

- [1] Barbieri B, Beverini N and Sasso A 1990 *Reviews of Modern Physics* **62** 603-44
- [2] Murnick D and Peer B J 1994 *Science* **263** 945-7
- [3] Murnick D, Dogru O and Ilkmen E 2008 *Analytical Chemistry* **80** 4820-4
- [4] Murnick D, Dogru O and Ilkmen E 2010 *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **268** 708-11
- [5] Tuniz C, Bird J, Fink D and Herzog C 1989 *Accelerator Mass Spectrometry, Ultrasensitive Analysis for Global Science* (Boca Raton: CRC Press)
- [6] Salehpour M, Håkansson K and Possnert G 2013 *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **294** 97-103
- [7] Lappin G, Noveck R and Burt T 2013 *Expert Opinion on Drug Metabolism & Toxicology* **9** 817-34
- [8] Eilers G, Persson A, Gustavsson C, Ryderfors L, Mukhtar E, Possnert G and Salehpour M 2013 *Radiocarbon* **55** 237-50
- [9] Persson A, Eilers G, Ryderfors L, Mukhtar E, Possnert G, Salehpour M 2013 *Analytical Chemistry* **85** 6790-8
- [10] Berglund M, Thornell G and Persson A 2013 *Journal of Applied Physics* **114** 033302

- [11] Berglund M, Grudén M, Thornell G and Persson A 2013 *Plasma Sources Science and Technology* **22** 055017
- [12] May R D and May P H 1986 *Review of Scientific Instruments* **57** 2242-5
- [13] Okil JO 2004 PhD thesis *Optogalvanic Spectroscopy for Atmospheric Carbon-Dioxide Concentration and Isotopic Ratio Measurements* Rutgers, NJ