

Measurement of positive gain on the 1315 nm transition of atomic iodine pumped by $O_2(a^1\Delta)$ produced in an electric discharge

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Laser action at 1315 nm on the $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ transition of atomic iodine is conventionally obtained by a near-resonant energy transfer from $O_2(a^1\Delta)$, which is produced using wet-solution chemistry. The system difficulties of chemically producing $O_2(a^1\Delta)$ has motivated investigations into gas phase methods to produce $O_2(a^1\Delta)$ using low-pressure electric discharges. In this letter we report on positive gain on the 1315 nm transition of atomic iodine where the $O_2(a^1\Delta)$ was produced in a flowing electric discharge. The electric discharge was followed by a continuously flowing supersonic cavity that was necessary to lower the temperature of the flow and shift the equilibrium of atomic iodine more in favor of the $I(^2P_{1/2})$ state. A tunable diode laser system capable of scanning the entire line shape of the (3,4) hyperfine transition of iodine provided the measurements of gain. © 2004 American Institute of Physics. [DOI: 10.1063/1.1784519]

The classic chemical oxygen-iodine laser (COIL) system¹ operates on the $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ electronic transition of the iodine atom at 1315 nm. The population inversion is produced by the near-resonant energy transfer between the metastable excited singlet oxygen molecule, $O_2(a^1\Delta)$ [denoted $O_2(^1\Delta)$ hereafter], and the iodine atom ground state $I(^2P_{3/2})$. Conventionally, the $O_2(^1\Delta)$ is produced by a liquid chemistry singlet oxygen generator (SOG). There are many system issues having to do with weight, safety, and the ability to rapidly modulate the production of the $O_2(^1\Delta)$, which have motivated investigations into methods to produce $O_2(^1\Delta)$ using flowing electric discharges. Early attempts to implement electric discharges to generate $O_2(^1\Delta)$ and transfer to iodine to make a laser by Zalesskii² and Fournier³ did not result in positive gain. Over the past several years, investigations into the possibility of a hybrid electrically powered oxygen-iodine laser have been performed with electric discharges to produce the $O_2(^1\Delta)$.⁴⁻⁹ These studies have shown that flowing electric discharges through oxygen containing mixtures, typically diluted with a rare gas, can produce significant quantities of $O_2(^1\Delta)$. Recent studies have demonstrated that $O_2(^1\Delta)$ yields greater than 15% using electric discharges,^{6,7,9} and modeling results^{4,7,8,10} have indicated that such a system may produce a viable laser. We report on the direct measurement of gain in atomic iodine resulting from electric discharge produced $O_2(^1\Delta)$.

In this letter, we report on measurements of positive gain on the $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ electronic transition of the iodine atom at 1315 nm pumped by resonance excitation transfer from $O_2(^1\Delta)$ produced in an electric discharge. A block diagram of the flow tube setup is shown in Fig. 1. A capacitive radio frequency (rf) discharge operating at 13.56 MHz was used as the excitation source. The plasma zone is approximately 4.9 cm in diameter and 25 cm long. Details of the

performance of the electric discharge can be found in Carroll *et al.*⁵

The supersonic diagnostic cavity is a Mach 2 nozzle with view port windows. The subsonic diagnostic duct has four windows through which simultaneous measurements are made of the optical emission from $O_2(^1\Delta)$ at 1268 nm, $I(^2P_{1/2})$ at 1315 nm, and the gain/absorption proportional to $[I(^2P_{1/2})] - 0.5[I(^2P_{3/2})]$. A Roper Scientific optical multi-channel analyzer (OMA-V) with a 512-element InGaAs LN₂ cooled array, attached to an Acton Research SP-150 monochromator, was used for the spectral measurements at 1268 and 1315 nm.

A variety of Micro-Motion CMF and Omega FMA mass flow meters were used to accurately measure the flow rates of the gases. An I₂ flow diagnostic, developed by Physical Sciences, Inc. (PSI), is based upon the continuum absorption of molecular iodine at 488 nm. The details of this diagnostic are described by Rawlins *et al.*¹¹ Pressure in the subsonic and supersonic flows regions was measured by capacitance manometers from MKS and Leybold.

Measurements of gain (or absorption) were made with the iodine-scan diagnostic (ISD), developed by PSI.¹² The ISD is a diode laser-based monitor for the small signal gain in iodine lasers. The system uses a single mode, tunable diode laser that is capable of accessing all six hyperfine components of the atomic iodine transition. It was calibrated in frequency for automated operation on the (3,4) hyperfine transition for our experiments. A fiber-optic cable was used

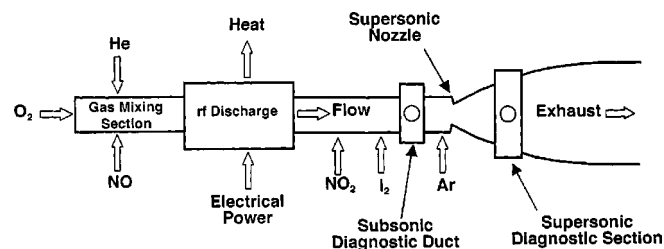


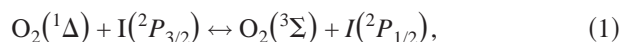
FIG. 1. Schematic of the experimental apparatus.

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to deliver the diode laser probe beam to the iodine diagnostic regions in the subsonic portion of the flow tube and in the supersonic cavity. Since the ISD uses a narrow-band diode laser, the measured line shapes can also be used to determine the local temperature from the Voigt profile.

Wedged antireflection coated windows were used on the sides of the cavity for the gain diagnostic to minimize etalon effects. A two-pass configuration (10 cm path length) was used in the subsonic section and a four-pass configuration (20 cm path length) in the supersonic section. $O_2(^1\Delta)$ yield measurements were made from the gain diagnostic and relative $I(^2P_{1/2})$ to $O_2(^1\Delta)$ spectral intensity measurements based upon the techniques originally developed by Hager¹³ and Davis and Rawlins.¹¹

During the course of this research, it was determined that electric discharge stability and temperature control are critical parameters to obtaining positive gain. Electric discharges sustained in moderate pressures (many to tens of Torr) of oxygen are prone to arcing and constriction. The discharge production of O atoms, O_3 , and other excited species adds higher levels of complexity to the downstream kinetics when the iodine donor species, which are not encountered in the purely chemical system, are added to the flow. The critical aspect of temperature control results from the equilibrium of the pumping reaction,



where the forward rate is 7.8×10^{-11} cm³/molecule s,¹⁴ and the backward rate is $1.04 \times 10^{-10} \exp(-403/T)$ cm³/molecule s,¹⁵ with the equilibrium rate constant ratio of the forward to backward reactions being $K_{eq} = 0.75 \exp(403/T)$,¹⁵ where T is the gas temperature. The threshold yield of $O_2(^1\Delta)$ for positive I^* to I inversion as a function of temperature can be written as $Y_{th} = 1/[1 + 1.5 \exp(403/T)]$.¹⁶ Note that the backward rate is slower, K_{eq} is larger, and Y_{th} is lower as T is decreased.

Several flow conditions were found that resulted in positive gain using the configuration shown in Fig. 1. A typical set of conditions are 4.0 mmol/s of O_2 mixed with 16.0 mmol/s of He and 0.2 mmol/s of NO flowing through a 400 W rf discharge. The discharge production of $O_2(^1\Delta)$ was enhanced by the addition of a small proportion of NO to lower the ionization threshold of the gas mixture. An additional 0.2 mmol/s of NO_2 was added downstream to scavenge some of the excess O atoms, followed by the injection of a secondary stream of 0.008 mmol/s of I_2 with 2.0 mmol/s of secondary He diluent. 20.0 mmol/s of Ar was injected further downstream to raise the pressure for better nozzle performance with our vacuum system. The pressures in the subsonic diagnostic duct and in the supersonic diagnostic cavity were 10.6 and 1.6 Torr, respectively.

Absorption in the subsonic region for these conditions was -0.009% cm⁻¹, with a temperature of 400 K, and an $O_2(^1\Delta)$ yield of approximately 15% (as computed from the gain technique outlined by Rawlins *et al.*¹¹). Based on these measurements, positive gain could be expected if the gas temperature could be sufficiently reduced. As such, gain measurements in the supersonic cavity were made for the above flow conditions as shown in Figs. 2 and 3. With 100 W of rf discharge power and a yield of approximately 5%, only absorption was observed. Upon raising the discharge power to 400 W, positive gain of $\approx 0.002\%$ cm⁻¹

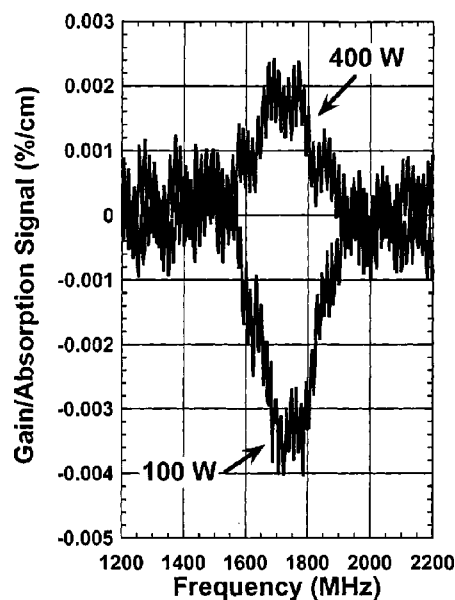


FIG. 2. Gain in the supersonic cavity as a function of frequency. Positive gain was observed at 400 W rf discharge power and absorption was observed at 100 W.

was measured. The measured line shapes indicate a temperature of ≈ 210 K at 100 W and ≈ 240 K at 400 W in the supersonic region. Over the parameter space investigated to date, the gain increases with power into the system; this fact gives encouragement for further increases in gain. Interestingly, the first measurement of positive gain on a classic liquid chemistry COIL system was also on the order of 0.001 %/cm.¹⁷

In conclusion, positive gain was measured on the $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ electronic transition of the iodine atom at 1315 nm pumped by a near-resonant energy transfer from $O_2(^1\Delta)$ produced in an electric discharge. A supersonic cavity was employed to lower the temperature of the flow and reduce the effect of the backward pumping reaction. This produced sufficient population inversion to observe a small, but easily identified positive gain of approximately 0.002 % cm⁻¹. The population inversion was linearly proportional to power deposition in the discharge. The critical is-

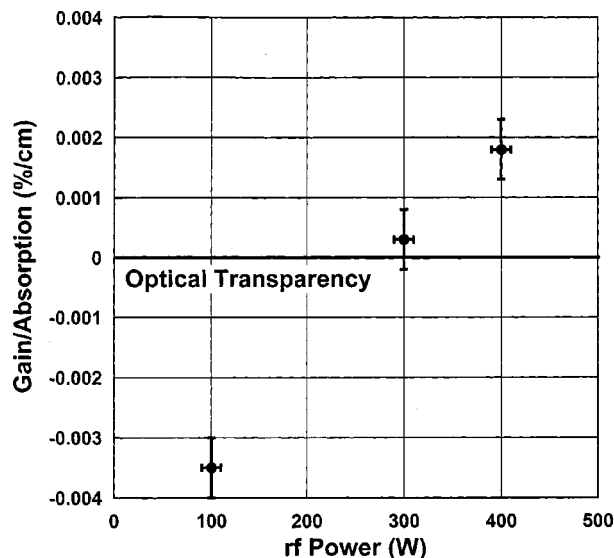


FIG. 3. Gain in the supersonic cavity as a function of rf discharge power.

sues which needed to be addressed to attain positive gain were improvements to the discharge flow and stability, along with a significantly more complete understanding of the gas phase kinetics.

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