Continuous-wave laser oscillation 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 difficulties in chemically producing $O_2(a^1\Delta)$ has motivated investigations into purely gas phase methods to produce $O_2(a^1\Delta)$ using low-pressure electric discharges. In this letter, we report on the demonstration of a continuous-wave laser on the 1315 nm transition of atomic iodine where the $O_2(a^1\Delta)$ used to pump the iodine was produced by a radio-frequency-excited electric discharge. The electric discharge was sustained in a He/O_2 gas mixture upstream of a supersonic cavity which is employed to lower the temperature of the continuous gas flow and shift the equilibrium of atomic iodine in favor of the $I(^2P_{1/2})$ state. The laser output power was 220 mW in a stable cavity composed of two 99.99% reflective mirrors.
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The classic chemical oxygen-iodine laser (COIL) system[1] 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 as $O_2(^1\Delta)$ 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. 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 Zalesskii[2] and Fournier[3] 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 $O_2(^1\Delta)$ yields greater than 15% using electric discharges,[6,7,9] and modeling results[4,7,8,10] have indicated that such a system can produce a viable laser. Recently, Carroll *et al.*[11] reported direct measurements of positive gain in atomic iodine resulting from electric discharge produced $O_2(^1\Delta)$.

In this letter, we report on the demonstration of a continuous-wave (cw) laser 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 radio-frequency (rf) electric discharge at 13.56 MHz operating between two internal hollow cathode electrodes was used as the excitation source. The plasma zone was 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 has a Mach 2 nozzle with windows that serve as view ports. 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 $[I(^2P_{1/2})] - 0.5 \cdot [(I^2P_{3/2})]$. A Roper Scientific optical multichannel analyzer (OMA-V) with a 512-element InGaAs LN_2 cooled array interfaced to an Acton Research SP-150 monochromator, was used for measurements at 1268 nm and 1315 nm.

Micro-Motion CMF and Omega FMA mass flow meters were used to measure the flow rates of the gases. The I_2 concentration was measured by a method developed by Physical Sciences Inc. (PSI) and is based on the continuum absorption of molecular iodine at 488 nm. Details of this diagnostic are described by Rawlins *et al.*[12] Pressure in the subsonic and supersonic flow regions were measured by capacitance manometers from MKS and Leybold.

Measurements of gain (or absorption) were made prior to running the apparatus as a laser using the iodine-scan diagnostic (ISD) developed by PSI.[13] 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. It was calibrated in frequency to enable automated operation for the (3,4) hyperfine transition for our experiments. A fiber optic cable was used 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, measurements of the lineshapes can also be used to determine the local temperature from the Voigt profile.

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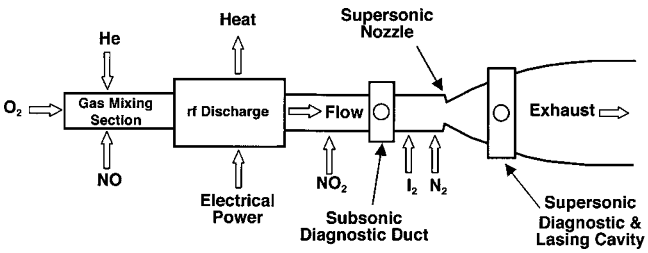

FIG. 1. Schematic of the experimental apparatus.

The windows on the sides of the cavity when using the gain diagnostic were wedged and antireflection coated 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) was used in the supersonic section. Measurements of the $O_2(^1\Delta)$ yield {defined as $Y = O_2(^1\Delta)/[O_2(^3\Sigma)+O_2(^1\Delta)]$} were obtained from the gain measurements, and the relative values of the spectral intensities measured for $I(^2P_{1/2})$ to $O_2(^1\Delta)$ using techniques originally developed by Hager,[14] Davis and Rawlins.[12]

Laser power measurements were made with a Scientech Astral™ model AC2500/AC25H calorimeter interfaced to a Scientech Vector™ model S310 readout, and were made at the same location in the supersonic laser cavity as were the gain measurements. The gain measurements were made first. The vacuum mirror mounts were then put in place for the laser power trials. Two mirrors with 1 m radius of curvature, provided by Los Gatos Research, Inc. (LGR), formed a stable optical cavity LGR characterized the mirrors by cavity-ring-down spectroscopy to have a reflectivity, R, of 99.987±0.001%. The mirrors were separated by approximately 38 cm. An infrared detection card from New Focus, Model 5842, with response between 800–1600 nm, was also used to observe the intensity profile of the beam.

Electric discharge stability and temperature control were found to be critical parameters to obtaining positive gain. Electric discharges sustained in moderate pressures (5–15 of Torr) of oxygen are prone to arcing and constriction. The production of O atoms, O_3, and other excited species by the discharge adds higher levels of complexity to the downstream kinetics when the iodine donor species are added to the flow. (These species are not usually encountered in the purely chemical system). The critical aspect of temperature control results from the equilibrium of the pumping reaction,

$$O_2(^1\Delta) + I(^2P_{3/2}) \leftrightarrow O_2(^3\Sigma) + I(^2P_{1/2}), \quad (1)$$

where the forward rate is 7.8×10^{-11} cm^3/molecule s,[15] and the backward rate is $1.04\times10^{-10}\exp(-403/T)$ cm^3/molecule s.[16] The equilibrium rate constant ratio of the forward to backward reactions is $K_{eq}=0.75\exp(403/T)]$,[16] where T is the gas temperature. The yield of $O_2(^1\Delta)$ for optical transparency as a function of temperature is $Y_{OT}=1/[1+1.5\exp(403/T)]$.[17] Note that the backward rate is slower, K_{eq} larger, and Y_{OT} lower as T is decreased.

The choice of mirror reflectivities was based on previous measurements of gain. For laser oscillations to occur, the gain coefficient at line center $\gamma_0(\nu_0)$ must satisfy

$$\gamma_0(\nu_0) \geq \frac{1}{2\ell_g}\ln\left(\frac{1}{R_1R_2}\right), \quad (2)$$

where ℓ_g is the length of the gain medium (5 cm for our experiment) and R_1 and R_2 are the mirror reflectivities. For similar flow conditions, we previously[18] obtained a gain of ≈0.005% cm^{-1}. With these values of gain and gain length, laser oscillation requires mirrors having reflectivity exceeding $R_1R_2=0.999\,50$, or $R_1=R_2=0.999\,75$.

Several flow conditions were investigated that resulted in positive gain and lasing using the configuration shown in Fig. 1.[11,18] The conditions used in these experiments were 3.0 mmol/s of O_2 mixed with 16.0 mmol/s of He and 0.15 mmol/s of NO flowing through the rf discharge. The discharge production of $O_2(^1\Delta)$ was enhanced by the addition of a small proportion of NO to lower the average ionization threshold and thereby also lower the sustaining value of E/N of the gas mixture. (NO has a lower ionization threshold than do O_2 and He, thus the addition of NO enhances the production of electrons, which increases the conductivity of the plasma, which reduces the electric field needed to sustain the plasma and, consequently, E/N is reduced). For these conditions, no additional NO_2 was employed to scavenge excess O atoms. (In earlier experiments that achieved positive gain using slightly different flow conditions, NO_2 was used to scavenge excess O atoms).[11,18] A secondary stream of ≈0.008 mmol/s of I_2 with 2.0 mmol/s of secondary He diluent was injected ≈63.5 cm downstream from the exit of the discharge. A tertiary flow of 55 mmol/s of cold N_2 gas (≈120 K) was injected further downstream to lower the temperature and to raise the pressure to improve the peformance of the nozzle with in our vacuum system. The pressures in the subsonic diagnostic duct and in the supersonic diagnostic cavity were 12.6 Torr and 1.55 Torr, respectively.

Gain for the above flow conditions at 450 W of rf discharge power is shown in Fig. 2 and peaks at 0.0067% cm^{-1} at line center. The line shape indicates a temperature of ≈180 K. The laser resonator was subsequently installed around the supersonic flow cavity and simultaneous measurements of $O_2(^1\Delta)$ yield (as computed from the gain technique outlined by Rawlins *et al.*)[12] and laser power were made as a function of rf discharge power as shown in Fig. 3. For the above flow conditions and 450 W rf power, a laser output power of 207 mW was obtained. The yield of $O_2(^1\Delta)$ was ≈17% with a temperature of ≈410 K in the subsonic diagnostic duct. [Note that the drop in $O_2(^1\Delta)$ signal beyond 400 W is believed to be a consequence of instabilities and thermal constriction that visibly develop in our existing dis-

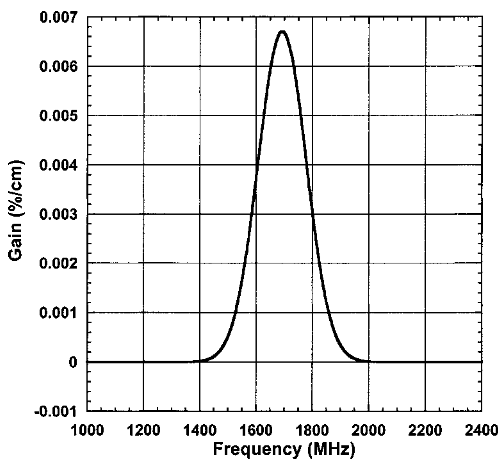

FIG. 2. Digitally filtered gain signal in the supersonic cavity as a function of frequency measured prior to lasing experiments.

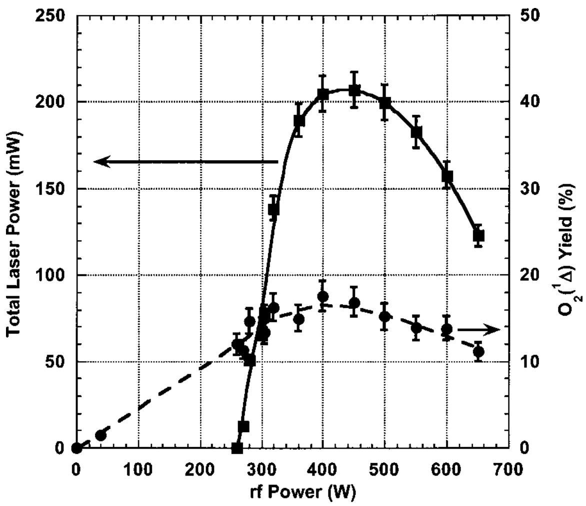

FIG. 3. $O_2(^1\Delta)$ yield in the subsonic diagnostic section and total laser power in the supersonic cavity as a function of rf discharge power.

charge under these flow conditions].[18] The beam shape was circular with a diameter of ≈1.9 cm, the same as the clear aperture of the mirror mounts.

The threshold for laser oscillation lasing occurs at 260 W of rf discharge power and an estimated $O_2(^1\Delta)$ yield of 12% in the subsonic flow tube, as shown in Fig. 3. Note that there is a roll off in laser power beyond 450 W that is greater than the drop in $O_2(^1\Delta)$ yield which is in part attributed to discharge instabilities. Even in the absence of discharge instabilities, laser oscillation would likely decrease at higher powers for these conditions as a consequence of two factors: (i) Higher powers result in higher gas temperatures and consequently lower gain, and (ii) progressively more O atoms are generated at higher powers while the NO flow rate was optimized for 450 W. [O atoms have been found to quench the excited $I(^2P_{1/2})$ atom].[18] The laser power was relatively stable at 220 mW±10 mW for more than 33 min, as shown in Fig. 4. The cause of the small oscillations in Fig. 4 with a period of approximately 33 s is unknown. For reference, the first measurement of laser action using a classic liquid chemistry COIL system produced 4 mW.[1]

In conclusion, cw laser action 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 shift the equilibrium of atomic iodine in favor of the $I(^2P_{1/2})$ state. This produced sufficient population inversion to observe positive gain of ≈0.0067% cm^{-1} followed by laser oscillations when two 99.99% reflectivity mirrors were used to form an optical resonator surrounding the gain medium. The laser output power was 220 mW and was stable for more than 30 min.

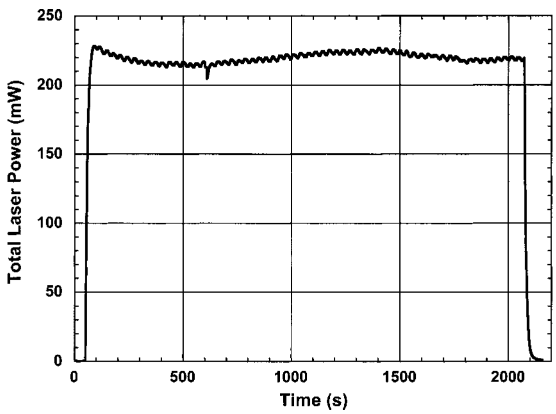

FIG. 4. cw laser power as a function of time.

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