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Historical Perspective of COIL

William E. McDermott
Chief Technology Officer
Directed Energy Solutions

ABSTRACT

The oxygen-iodine laser was the first electronic transition chemical laser. It first lased 25 years ago at the Air Force Weapons Laboratory (now the Air Force Research Laboratory). The development started several years earlier and involved the support of many people in the laser community. I would like to describe the early thoughts, insights and even misconceptions that we had in the early days. I will also highlight the contributions of many of the people and organizations that contributed to the early development of the COIL laser.

1. A Brief Background on $O_2(^1\Delta_g)$

Chemical sources of $O_2(^1\Delta_g)$ have been known for many years. Mallet [1] first reported the red glow obtained when hypochlorite solution and hydrogen peroxide was mixed. The $O_2(^1\Delta_g)$ molecule was first identified by Herzberg in the spectrum of the sun in 1934. Groh [2] mentions the red chemiluminescence obtained when gaseous chlorine or bromine is added to basic hydrogen peroxide. Later, Groh & Kirriman [3] mixed gaseous chlorine with KOH and hydrogen peroxide and proposed that the red glow is from two $O_2(^1\Delta_g)$ molecules colliding. Later, Seliger [4] published the spectra of the hypochlorite – peroxide reaction in the red at 634 nm. For me, the seminal article on excited oxygen was Khan and Kasha [5]. Even though the chemical process that produced a metastable electronically excited species was known, the marriage of oxygen and iodine came in discharge studies.

2. Iodine and $O_2(^1\Delta_g)$

The reaction of iodine and discharged oxygen was first reported by Ogryzlo and his group [6]. They observed that when iodine was added to excited oxygen, a bright yellow glow was seen. This was recognized as I_2 B state emission. They also observed a strong emission at 1.315 μ, the iodine atom $^2P_{1/2} - ^2P_{3/2}$ transition. The excitation of the iodine was attributed to the near resonant pumping by $O_2(^1\Delta_g)$. The iodine atoms were thought to be formed by the dissociation of molecular iodine by $O_2(^1\Sigma_u)$. This work didn't attract the attention of the laser community until later. In 1969, Elmer Ogryzlo went on sabbatical to Brian Thrush's laboratory in England. There he suggested the oxygen-iodine system as an interesting system for Thrush's graduate student, R. G. Derwent. The suggestion generated several papers [7], [8], [9], [10], and [11]. In their third paper, they suggested that an inversion could be achieved on the iodine atom $^2P_{1/2} - ^2P_{3/2}$ transition if a sufficient $O_2(^1\Delta_g)$ fraction could be produced. Using the equilibrium constant for the energy pumping reaction, They showed a flow containing about 15% $O_2(^1\Delta_g)$ could produce an inversion on the iodine $^2P_{1/2} - ^2P_{3/2}$ transition. It was radical to propose an equilibrium as a pumping reaction – in a two level system terminating in the ground state!

$$I(^2P_{\frac{3}{2}}) + O_2(^1\Delta_g) \Leftrightarrow I^*(^2P_{\frac{1}{2}}) + O_2(^3\Sigma_u) + \Delta E$$

$$\Delta E = 279 \text{ cm}^{-1}$$

$$K_{eq} = 0.75 e^{\frac{401.42}{T}}$$

$$\alpha = \sigma_0 \left(\left[I^*(^2P_{\frac{1}{2}}) \right] - \frac{1}{2} \left[I(^2P_{\frac{3}{2}}) \right] \right)$$

$\alpha > 0$ for

$$\frac{O_2(^1\Delta_g)}{O_2(^1\Delta_g) + O_2(^3\Sigma_u)} > 14.5\%$$

at T = 300K

Gas and Chemical Lasers and Intense Beam Applications III, Steven J. Davis, Michael C. Heaven, Editors, Proceedings of SPIE Vol. 4631 (2002) © 2002 SPIE · 0277-786X/02/$15.00

3. The Search for a Chemical Source of $O_2(^1\Delta_g)$

This paper produced a considerable amount of interest at the Air Force Weapons Laboratory (AFWL); who, like many other research organizations, had developed an interest in short wavelength chemical lasers. The first advocate was Dr. Al MacKnight who brought the Derwent and Thrush article to my attention. I had just arrived at the Air Force Academy (USAFA), where I was teaching Instrumental Analysis in the Chemistry Department. I had been selected to spend the next year as an exchange officer to the Frank J. Sieler Research Laboratory (FJSRL) – a part of the Air Force Office of Scientific Research (AFOSR) – and was looking for research projects of interest to the Air Force. I had visited the AFWL the previous year and met a number of the technical people there. In the summer of 1973, I began working on electronic transition chemical lasers. The initial funding for the effort was $10,000 provided by Dr. Don Ball, then head of AFOSR. This money went towards purchase of an S-1 photomultiplier and electronic data recording equipment. I began to look at a number of systems including the Ba-N_2O-CO system, the reaction of SnH_4 with N_2O, and various alkali metal – oxidant systems. These were experiments in which the species were burnt in a flame or mixed in the afterglow of a microwave discharge and the spectra recorded. Nothing much of interest came from these experiments. In those days, we didn't have the Internet to browse; so I would spend hours in the library tracking down references. I was aware of the hypochlorite – peroxide method of producing $O_2(^1\Delta_g)$ since Al MacKnight was doing experiments with that system as well as microwave discharges. One day – I think it was in mid-1974 – I ran across the Khan and Kasha article. Michael Kasha was doing biomedical research on the possible role of $O_2(^1\Delta_g)$ in cancer. He had just gotten a new graduate student – A.U. Khan. Michael told me later that he wanted to see what this new guy could do in the lab, so he gave him the problem to record the spectrum of $O_2(^1\Delta_g)$. About a week later, he came back with the spectrum. Khan had built a small bubbler shaped somewhat like a saxophone (hence the name "Hirtzian device" – after Al Hirt, a famous New Orleans jazz musician) with a fritted disk at the end. He bubbled chlorine gas through a basic solution of hydrogen peroxide and recorded the spectra. In the article that I saw, a print of the spectrum of the emission in the 1.27μ region was attached. It had been recorded on photographic film at good resolution. The text did not mention what I saw in the spectrum. It was clearly rotationally resolved! There seemed to be no continuous emission between the rotational lines. Since molecules in solution are strongly perturbed during emission by collisions, emission from molecules in solution will not show rotational structure. The emission Ahsan Khan saw was from the gas phase. At that point, I was convinced that the liquid phase reaction between chlorine and basic hydrogen peroxide could produce high yields of gaseous $O_2(^1\Delta_g)$.

4. Early Research on Chemical Sources

4.1. Air Force Weapons Laboratory (1973 – 1977)

At the AFWL, Al MacKnight pursued a number of approaches to establish oxygen-iodine as a laser. First, a number of flow studies were done and several attempts were made to lase using microwave excited oxygen as a source. None of the laser experiments were successful. MacKnight also tried to react gaseous hydrogen peroxide with calcium hypochlorite to produce $O_2(^1\Delta_g)$ which was also unsuccessful. The AFWL did, however, support a number of research projects to develop a COIL laser. It also encouraged AFOSR to support additional University research such as my effort at the USAFA.

4.2. University of California, Santa Barbara (1975-1977)

At the Quantum Institute at the University of California at Santa Barbara, Paul Lee and Warren Slafer attempted to build a chemical generator using bromine or chlorine condensed on a rotating dewar. The frozen halogen was then sprayed with basic hydrogen peroxide. It was reported to work adequately (~25%) and was delivered to the AFWL in early 1977. It didn't seem to catch the interest of the researchers there at the time.

4.3. *Rockwell Science Center (1976-1978)*

A more important effort was started at Rockwell Science Center by Bob Coombe and his collaborators – A. T. Pritt, I. B. Goldberg, D. Pilipovich, and R. I. Wagner. The investigation centered on the use of chlorine fluorosulfate ($ClOSO_2F$) and BHP. The effort was less important in the details of the chemical generation process and more important that it produced the first measurement of gain in a chemical pumped iodine system [12]. We discovered later that chlorine fluorosulfate and BHP generates chlorine gas which then reacts with the excess BHP to produce $O_2(^1\Delta_g)$. Ira Goldberg also recognized the value of EPR as a measurement standard for excited oxygen and used it in these studies.

4.4. *Air Force Academy (1973-1976)*

After reading the Kasha and Khan article, I embarked on devising a way to use the reaction between chlorine and BHP to produce gaseous $O_2(^1\Delta_g)$. I reasoned that the control of the gas-liquid interface was important and that the reaction would have no hope of producing a high yield of excited oxygen unless the reaction rate of chlorine with BHP was faster than the quenching rate of $O_2(^1\Delta_g)$ on BHP. The first experiment was to determine the rate of reaction of chlorine with BHP. Since funding was limited, we couldn't look at the disappearance of chlorine as it reacted with BHP; so we studied the reaction of chlorine with NaOH. We could follow that reaction with a pressure gauge. Chemically, these reactions should be about equal in rate. A recording mercury manometer was devised (we couldn't afford a capacitance manometer) to measure the pressure drop as chlorine passed over concentrated NaOH solution. John Viola, who was also teaching at USAFA, was a great help in designing and running these experiments. The apparatus is described in a note [13]. We found that the sticking coefficient of chlorine with NaOH was about 10^{-2}. We reasoned that the quenching coefficient of $O_2(^1\Delta_g)$ on BHP would be less than 10^{-5}, therefore it appeared that the system would work. The real question in my mind was how we could accurately measure the yield of excited oxygen relative to the ground state. I gave a seminar to the Physics Department in 1974 and mentioned the lack of a good method to measure the excited oxygen yield. At the end of the seminar, one of the Physics instructors, Dave Thomas, came up to me and suggested that gas phase electron paramagnetic resonance (EPR) could measure both ground state and excited oxygen. Even more important, there was an EPR in one of the Physics laboratories. With Dave's help, we soon showed that we could measure both species accurately. At this point, we built a small rotating wetted wall generator that we could attach to the EPR. It is shown schematically in Figure 1.

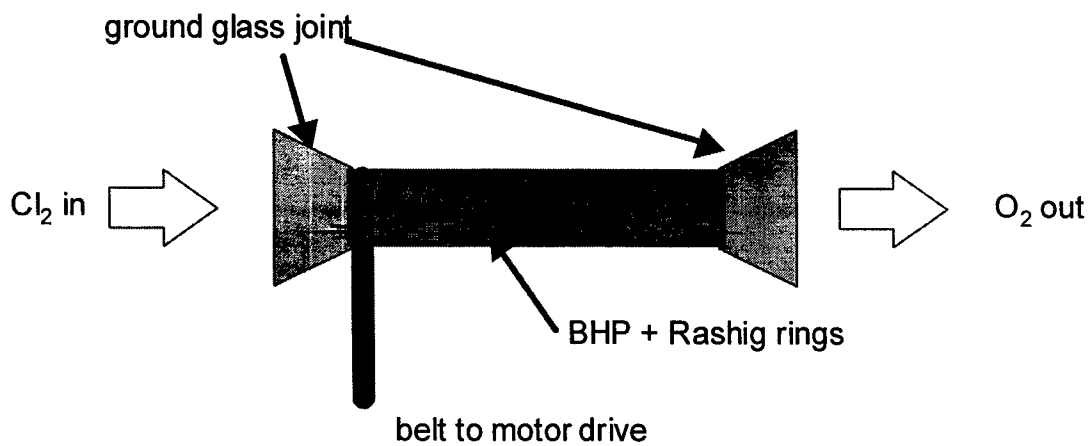

Figure 1 The first >15% chemical generator

Among the experiments I was able to do while at USAFA was observation on the EPR of $^2P_{1/2}$ iodine atoms excited by the chemical generator. This showed that the water produced by the reaction did not stop the

iodine dissociation process. The performance, a yield greater than 15% at the generator, was reported at the Second Summer Colloquium on Electronic Transition Lasers [14]. By then, I had left the USAFA and was attending a one-year Air Force middle management school. I was able to attend the conference because AFOSR provided funds. The paper did not generate any interest in the community.

It was extremely important during this early period to be able to discuss the problems of chemical lasers with many of the experts in the field. AFOSR and AFWL funding gave the researchers an opportunity to interact with many of the best scientists of the 20th century. Herb Broida and Dick Zare come to mind as people who were always available to help in anyway they could. Elmer Ogryzlo and J.C. Polyani both came to the USAFA under AFOSR funding to discuss electronic transition chemical lasers with me.

5. Early COIL Work at AFWL (1977 – 1981)

On arriving at AFWL in late 1977, there was already a well-equipped laboratory. It contained a flow system, laser cavity and EPR system. It had been configured to test the generator developed at RSC. I inherited the iodine laser team, which consisted of Nick Pchelkin, Dave Benard and a couple of technicians. Down the hall, I had access to one of the best experimental physicists in the world – Steve Davis. I had turned down Leroy Wilson's offer to work on a "real, short term, high payoff' system called ALFA. The first few months were occupied with two problems. The first was that the "system" model developed at AFWL predicted that more than 25% $O_2(^1\Delta_g)$ was required to achieve threshold. Any generator had to beat this new, higher standard. In fact, I was able to show that the model incorrectly calculated the equilibrium constant and that Derwent and Thrush were right after all [15]. The second problem was an article by two scientists at the Naval Research Laboratory who measured the branching ratio between the pumping reaction and the non-resonant branch [16]. The experiment used photodissociation of alkyl iodides as a source of $I(^2P_{1/2})$. Their value was 40%. Clearly, if that were true, there would never be a sustained inversion. I never could figure out what they did wrong. I knew their answer was inconsistent with the experiments I did at USAFA and Dave Benard's measurement of gain at RSC. Always believe the experiment that most closely duplicates what you are trying to do.

By this time, the management was getting upset with the "slow" pace of our effort to get the CFS generator running and directed me to stop advocating the chlorine – BHP reaction and immediately install a CFS system on the test stand. Being a good soldier, I called Nick Pchelkin in and asked him how long it would take to get a chlorine line plumbed into the lab. His answer was 8 AM tomorrow morning – I told him to do it. I then had a short meeting with Nick and Dave Benard to figure out what generator to start testing with. I advocated my rotating, wetted wall device. Nick didn't like that because it would take too long to build. I described the Khan and Kasha experiment and Nick was pretty excited because he could have a bubbler ready by the next day. I didn't think a bubbler would work because we didn't have a direct control of the gas-liquid interface. But you never know until you try the experiment, so I told Nick to go ahead with the bubbler. It is shown in Figure 2. We measured over 35% $O_2(^1\Delta_g)$ on the EPR the first time we turned on the device. In the experiment, I found I was able to control the gas-liquid interface by a combination of liquid depth and flow rate. In fact, the term bubbler was a misnomer. I really pumped the chlorine through the liquid at a very high rate – the flow formed conical channels and the liquid was pretty violently agitated. Chemical engineers call that a sparger.

At this point, lasing was not far off. As usual, we were still having trouble getting enough iodine into the laser, so Dave Benard redesigned the iodine source. Nick Pchelkin also rigged up a $O_2(^1\Delta_g)$ meter using a pressure tap and the photometric detector we had cross calibrated with the EPR. We were not using diluent so pressure was a measurement of total oxygen and chlorine. The "yield" was really $O_2(^1\Delta_g)$ divided by chlorine in. The detector was at the exit of the laser cavity. Dave suggested the most important diagnostic. Two IR detectors with narrow band 1.315μ filters were placed on the laser cavity – one looking down the optical axis; the second on the side perpendicular to the laser axis. The latter measured spontaneous emission and the former laser emission. Nick had these fed to an X-Y recorder. As $O_2(^1\Delta_g)$ was added before the mirrors were aligned, the two signals tracked linearly. As we tweaked the mirrors, we could recognize a positive change by an increase in the end emission as the side emission stayed constant. On 1 Dec 1976, the end emission suddenly shot off scale for the first time announcing laser action. We also had an IR phosphor

card. It showed a distinct higher order transverse mode. It looked like the 2,6 Hermite-gaussian mode shown on page 688 of Siegman [17]. The $O_2(^1\Delta_g)$ meter normally oscillated between 35% and 40%; but when the laser turned on, it immediately dropped to a rock constant 15%. During the several minutes of 4 mW lasing, the mode actually jumped several times. It was a pretty exciting day [18].

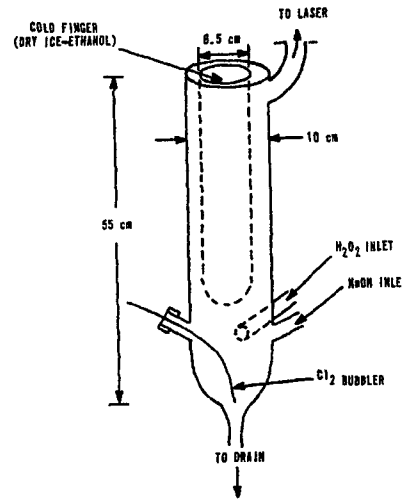

Figure 2 The first oxygen generator to power a laser.

That evening, Dave and Nick went out to dinner and designed the next laser on a napkin while waiting for their steak to be delivered. I scaled up the generator to match the transverse flow channel the next day. It would have been ready in a week if we had been able to get the laser built outside, but we were required to use the base model shop which took 6 months to build the laser cavity. In the meantime, Dave borrowed a small transverse flow cavity from Johns Hopkins that we used for flow studies. This was called COIL II. The second laser was called COIL III for that reason. It produced over 100 watts – a 25,000 times scale up [19].

The next few years were an exciting time. We had tremendous support from Pete Avizonis, the AFWL Chief Scientist, who became – and remains to this day – an ardent advocate of the COIL laser. Rick Heidner contributed the first systematic study of iodine dissociation [21]. The results were not always positive. Rick's work, coupled with some ab inito calculations done by Harvey Michaels; suggested that dissociation did not occur from the $O_2(^1\Sigma)$ channel, but through vibrationally excited molecular iodine. Quenching of the intermediate state could make scaling to higher pressures impossible. Steve Davis then began critical work to unravel this new dissociation mechanism [20]. Bob Shea took over the computer model and continually reminded me that our understanding of the kinetics was limited - motivating me to make sure we supported as many kinetic studies as we could. He was also the first to calculate the power in the flow and identify pressure scaling problems at subsonic velocity. Verne Schlie developed a gain probe for the COIL laser – a CW photolysis laser [22]. This was the first example of international cooperation – Verne spent a summer at the Max Planck Institute at Garching, Germany learning the details of photolysis lasers from the experts there. Verne also pointed out that there might be a problem with anomalous dispersion in COIL. This formed an interesting Ph.D. thesis topic for an Air Force Institute of Technology (AFIT) student named Dave Neumann. Dave and Steve Davis developed a method to measure the broadening coefficient of $^2P_{1/2}$ by oxygen and show that anomalous dispersion would not be a problem. On finishing his degree; Dave joined our little; but growing, group. He was to have a major impact on COIL development.

Not all of our effort was confined to in-house work. As a matter of fact, we gave funding to almost anyone who was interested in working on COIL lasers. Rocketdyne, Garrett AirResearch, McDonnell-Douglas Research Laboratories, and Bell Aerospace all worked on developing and understanding generators. Each also tried a laser – some worked and some didn't. We learned as much from the failures as we did from the

successes. Wayne Soloman ran the effort at Bell and is still active in the field at the University of Illinois. Al MacKnight was the leader at Garrett. I also felt that the McDonnell-Douglas effort (Ralph Richardson, Harvey Lilenfeld and Charlie Wiswall) provided key technical data [23]. Orv Sandall from the University of California at Santa Barbara, a consultant to Rocketdyne, gave us the first real chemical engineering analyses of the generator [24]. Steve Hurlock led the experimental team at Rocketdyne providing the data. Following that round, TRW and Bell Aerospace looked at characterizing and scaling the laser. I've already mentioned the work of Rick Heidner at Aerospace Corporation who continued to make major contributions to COIL kinetics. Jim Hurst at the Oregon Graduate Center made the first detailed study of the liquid phase chemistry [25]. He concluded that the intrinsic yield of $O_2(^1\Delta_g)$ in solution was near 100%.

At AFWL, we continued our scale-up efforts designing and building a multi-kilowatt subsonic laser. I left the AFWL just as that device was coming on line; leaving Gordan Hager who we had just hired from Rocketdyne and Doug Loverro, an ex-student of mine from USAFA, with the problem of getting it to work – which they did [26]. Gordan is still at the AFWL, although he is concentrating on his successful laser – the second electronic transition chemical laser, NCI-I. Bob Coombe also played an important role in the NCI-I discovery. During the same period, we had to face the problems of scaling subsonic devices – they would be large and probably exhibit poor beam quality. I asked Dave Neumann to head a technical group to develop a recommendation on the future course of COIL development. His recommendation to develop a supersonic COIL laser was critical to the current success of COIL. At that time it was quite a risk, a previous attempt had failed and no one had ever pushed $O_2(^1\Delta_g)$ through a supersonic nozzle. The proper place to inject the iodine and indeed the behavior of iodine dissociation at high pressures was a real unknown. Dave did succeed in demonstrating the first supersonic COIL laser about a year later [26]. About this time, Keith Truesdell also joined us from Rocketdyne. Keith remains one of the major drivers in COIL development.

6. An Explosion of Research

By the early 1980's, a number of groups began to exploit the potential of the COIL laser. Lasers were built in Russia, Israel, Japan, France, China, Germany and the Czech Republic. I have listed some of these lasers and their characteristics in Table 1. Some of the major advances were made during this time period. I've already mentioned Dave Neumann's successful supersonic laser. Marciel Zagidullin's invention of the jet generator was another major step [27]. Sani Yoshida's demonstration of greater than 40% laser efficiency showed COIL could be engineered to very high efficiency. Gordan Hager continued his innovative work by both frequency doubling [28]and Q-switching COIL [29]. The technical detail of COIL laser development can be found in the outstanding review by Keith Truesdell, Charlie Helms, and Gordon Hager [26]. Charlie Helms also did exceptional work on laser efficiency and mixing nozzles. I would also be amiss if I didn't mention the extraordinary work done by Mike Heaven to unravel the mystery of iodine dissociation [30], [31]. Mike will follow this address and talk in detail about his current research. We also will hear from many of the people who have contributed to COIL and I will not attempt to cover their many contributions here. I would like to highlight the importance of diagnostics – Steve Davis has been the leader in this most important effort. We will also hear about some of the commercial applications of COIL in the material processing industry. Another major application – The Airborne Laser – was reviewed yesterday by Steve Lamberson.

7. Conclusion

I would like to highlight the contributions of a few organizations whose contributions to the early development of COIL has been often overlooked. The AFOSR sponsored research in the mid-1970's was absolutely critical to the invention of COIL. Not only did it sponsor my own first efforts to develop a chemical generator, but supported technical efforts throughout the world. The AFIT also supported development by educating many of the key scientists involved in the US COIL effort. My Ph.D. was supported through the AFIT Civilian Institute Division as was my roommate – Greg Canavan, who ran the ARPA Electronic Laser program in the late 1970's. Graduates of the AFIT residence program included Nick Pchelkin and Dave Neumann. The

importance of a broad-based support for fundamental research and scientific education cannot be underestimated.

As a final note, I would like to assure you that advances in electronic transition lasers will continue to occur. Gordon Hager's session on NCl – pumped iodine is one example. At Directed Energy Solutions, I've been working with Dave Neumann on a new source of high pressure, high yield $O_2(^1\Delta_g)$. Initial experiments have produced excited oxygen concentrations in excess of an atmosphere. Such a source would reduce the weight and volume of a COIL laser by a factor of ten.

Year	Ref	Organization	Cl2 Flow	Power	Delta	Efficiency	Comments
			moles/s	watts	w/cm2		
		USA					
1977	[18]	AFWL	0.004	0.004	2.00E-05	1.00E-05	longitudinal flow, subsonic
1978	[32]	AFWL	0.03	150	1.2	5%	subsonic
1982	[26]	AFWL	0.60	4600	4.6	8%	subsonic
1984	[26]	AFWL	0.15	1600	40	12%	supersonic
1989	[26]	AFWL	1.80	39000	172	24%	supersonic, ROTOCOIL
1989	[28]	AFWL					frequency doubled
1990	[29]	AFWL		630			magnetically Q switched
1991	[26]	AFPL	0.50	10000	200	22%	supersonic, RADICL
1996	[33]	AFPL	0.07	1750	197	28%	compact supersonic, VERTICOIL
1996	[34]	AFPL					mode locked COIL
1979	[23]	McD	0.02	10	0.08	1%	subsonic
1984	[35]	McD	0.02	180	0.10	10%	subsonic
1985	[36]	McD					
1981	[26]	TRW	0.20	2000	7	11%	subsonic
1984	[26]	TRW	0.30	4200	84	15%	supersonic
1981	[26]	RD	0.05	150	0.9	3%	subsonic
1985	[26]	RD	0.15	1900	58	14%	supersonic
1995	[37]	RD	0.90	17500	400	21%	supersonic, mode limited - aperture
1995	[37]	RD	0.50	13716	258	30%	supersonic
		RUSSIA					
1982	[38]	VNIIEF		0.01			subsonic
1983	[38]	VNIIEF		180			subsonic
1986	[38]	VNIIEF		900			subsonic
1990	[38]	VNIIEF		4000			subsonic
1984	[39]	Lebedev, Moscow	0.001	5			subsonic, 50% H2O2
1989	[40]	Lebedev, Moscow					photodissociation of ozone
1991	[41]	Lebedev, Samara					jet generator
1997	[42]	Lebedev, Samara	0.01	200		22%	jet generator
		Israel					
1982	[43]	Ben Gurion U		5			subsonic
1994	[44]	Ben Gurion U		9			supersonic
1997	[45]	Ben Gurion U	0.01	177		18%	supersonic
		Japan					
1983	[46]	Nat Def Acad		10			subsonic
1987	[47]	Laser Lab, Chiba	0.002	40		21%	subsonic
1989	[48]	Laser Lab, Chiba	0.005	200		41%	subsonic
1989	[49]	Laser Lab, Chiba	0.07	1020		16%	subsonic

Year	Ref	Organization	Cl2 Flow	Power	Delta	Efficiency	Comments
		France					
1984	[50]	ONERA		4			subsonic, 30% H2O2
1991	[51]	ONERA		570			
		China					
1988	[52]	Dalian Inst Chem Phys					electrical discharge, 130 mJ
1994	[53]	Dalian Inst Chem Phys	0.03	178		7%	
1995	[54]	Dalian Inst Chem Phys	0.15	1000		7%	supersonic
1996	[55]	Dalian Inst Chem Phys	0.30	5000		18%	supersonic
		Czech Rep					
1991	[56]	Inst of Physics	0.01	58		5%	subsonic
		Germany					
1997	[57]	DLR		5000			generator from AFPL

Table 1 Selected COIL Laser Devices

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