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Innovative Lasers for Uranium Isotope Separation

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ABSTRACT

Copper vapor laser have important applications to uranium atomic vapor laser isotope separation (AVLIS). We have investigated two innovative methods of exciting/pumping copper vapor lasers which have the potential to improve the efficiency and scaling of large laser systems used in uranium isotope separation. Experimental research has focused on the laser discharge kinetics of 1) microwave, and 2) electron beam excitation/pumping of large-volume copper vapor lasers.

Microwave resonant cavity produced copper vapor plasmas at 2.45 GHz, have been investigated in three separate experimental configurations. The first examined the application of CW (0-500W) power and was found to be an excellent method for producing an atomic copper vapor from copper chloride. The second used a pulsed (5kW, 0.5 - 5 kHz) signal superimposed on the CW signal to attempt to produce vaporization, dissociation and excitation to the laser states. Enhanced emission of the optical radiation was observed but power densities were found to be too low to achieve lasing. In a third experiment we attempted to increase the applied power by using a high power magnetron to produce 100 kW of pulsed power. Unfortunately, difficulties with the magnetron power supply were encountered leaving inconclusive results. Detailed modeling of the electromagnetics of the system were found to match the diagnostics results well.

An electron beam pumped copper vapor system (350 kV, 1.0 kA, 300 ns) was investigated in three separate copper chloride heating systems, external chamber, externally heated chamber and an internally heated chamber. Since atomic copper spectral lines were not observed, it is assumed that a single pulse accelerator is not capable of both dissociating the copper chloride and exciting atomic copper and a repetitively pulsed electron beam generator is needed.

A. INTRODUCTION AND BACKGROUND

Atomic Vapor Laser Isotope Separation (AVLIS) is an important new method of producing enriched uranium. It is based upon the principle that ^{235}U and ^{238}U absorb different wavelengths of visible light. A copper vapor laser (CVL) is used to excite/pump dye lasers which generate the specific wavelengths needed to photoionize ^{235}U atoms in a vapor stream of uranium. These ^{235}U photo-ions are then deflected by electromagnetic fields onto a collector, whereas ^{238}U atoms pass straight through the system, unaffected. The high selectivity of the AVLIS method provides large efficiencies (greater than 10,000), unlike gaseous diffusion and gas centrifuge techniques which rely on isotopic mass differences and result in low enrichment efficiencies (0.01). The combination of high enrichment efficiency, low capital, low energy and low operating costs, make AVLIS an attractive, economic alternative to traditional separation techniques. The Department of Energy has funded a Laser Demonstration Facility which is currently developing the AVLIS technique at Lawrence Livermore National Laboratory (LLNL).

The Department of Energy has chosen copper vapor lasers (CVLs) as their "baseline technology"¹ for AVLIS because CVLs have the optimal properties for this method. Advantages of copper vapor lasers are: 1) copper vapor lasers can be driven at high repetition rates (4 kHz) which are needed to maintain large efficiencies, 2) CVLs have short pulse lengths (200 ns) needed to provide photoionization before relaxation can occur, 3) CVLs produce large average powers necessary to separate large amounts of materials, and 4) laser isotope separation CVLs produce narrow band light at the needed wavelengths. Copper vapor lasers currently used in the DOE AVLIS facility at LLNL are driven by repetitively pulsed electrical discharges between electrodes located near the ends of the insulating laser tube. However, electrical discharge type of excitation schemes have a number of technical problems which need to be solved.

There are several important shortcomings of the pulsed discharge copper vapor lasers currently used in the AVLIS program at LLNL. In order to achieve economies of

scale in laser isotope separation, it is required that copper vapor lasers are scalable to large volumes and powers. However, there has been some concern^{1,2} over the physical limitations of the size of the electrical discharge chambers because large-diameter copper vapor lasers exhibit a "plasma skin effect" which can delay the penetration of the applied electric field. Plasma skin effects can cause hollow laser beam intensity profiles. Furthermore the length of electrical discharge¹ cannot be extended indefinitely because of the limitations in high-voltage pulsed power technology.

During the past three years, we have initiated an investigation of two, new, innovative methods of excitation/pumping copper vapor lasers which will enhance their applicability to isotope separation and may overcome the shortcomings of the conventional electrical discharge excitation/pumping method:

- 1) Microwave excitation/pumping, and
- 2) Electron beam excitation/pumping.

The advantages to using microwaves for laser excitation/pumping: 1) Microwave energy can be very efficiently coupled to the gas. We have achieved efficiencies of greater than 95% in helium-copper chloride discharges. 2) high power, repetitively pulsed microwave technology is well established from radar work and is relatively inexpensive, particularly at 2.45 GHz, 3) microwave systems are scalable to high powers (several MW commercially available magnetrons), 4) the pulse rate needed for the application to isotope separation (several kHz) are well within the range of commercially available microwave systems, and 5) waveguide transmission of microwaves from the source to the load is reliable, inexpensive, and does not add inductance as would a high voltage cable. An additional feature of microwave discharges is the absence of electrodes, so electrode contamination of the laser medium and degradation of the electrodes and laser windows are eliminated. Note the only previous studies of microwave pumped lasers have centered around rare-gas halogen excimer lasers³⁻⁷,

ArXe⁸, HeNe⁹ and argon¹⁰. Very recently a Scottish research group has reported waveguide based high power (MW) microwave pumping of copper halides.¹¹

Electron beam excitation would have a number of advantages in pumping large copper vapor lasers for atomic vapor laser isotope separation: 1) Extremely large laser gas volumes can be excited, since the electron range can be very large, 2) electron beam excitation does not exhibit a plasma skin effect as does electrical discharge pumping, 3) electron beam excitation produces very uniform laser output power profiles for large area beams, 4) electron beams can deposit a larger amount of energy to the system (by two or three orders of magnitude) compared to the few joules deposited by the electrical discharges,¹² 5) electron beam excitation does not require the use of electrodes which can degrade and cause contamination, and 6) electron beam excitation is very efficient in converting electron beam energy to laser output energy. Borovich et al.¹² estimate that 3% of the electron beam energy could result in laser energy from an electron beam pumped copper vapor laser.

Details of the research performed for this project are given in the following sections and in the publications attached in the appendix.

B. MICROWAVE PROGRAM

The experimental system consisted of a microwave circuit for transmitting and measuring 2.45 GHz microwave power, both pulsed and CW, a right angle cylindrical Asmussen resonant cavity for transferring the microwave power to the laser gas discharge and a quartz tube for containing the gas mixtures. A quartz tube, 2.54 cm in diameter, was placed along the center axis of the microwave cavity and contained the discharge. The quartz tube (melting temperature ~2000K) was surrounded by glass wool in an attempt to keep the wall temperature close to the discharge temperature and to reduce the temperature gradients. See Figure 1 for a schematic of this system.

The Asmussen cavity consisted of a 17.8 cm diameter right cylindrical cavity made of brass, with a sliding short which allowed for adjustable cavity lengths from 6 to 16 cm.¹³ The sliding short and the excitation probe were adjusted for each operating point so that the incident power was maximized and the reflected power was minimized. This cavity is designed to excited various electromagnetic modes at 2.45 GHz depending upon the position of the sliding short and probe position. Because of the simplifying electromagnetics and the ease of operation, the cavity was always used in the TM₀₁₂ mode.

Initial diagnostic development was performed on air discharges and the details are given in the paper "Microwave Resonant-Cavity Produced Air Discharges" published in the IEEE Trans. on Plasma Sci., Vol 19, No. 2 April, 1991. See the appendix for a copy of this paper. The diagnostics mainly centered on optical emissions spectroscopy as well as some modeling of the electromagnetics.

The first set of experiments using copper chloride concentrated on producing copper atomic vapor in a CW microwave plasma. First we tried heating an external reservoir of copper chloride upstream of the discharge region. We found that the copper tended to recombine on the walls of the quartz tube before reaching the discharge region. We found that placing copper chloride directly in the discharge tube worked much better. The discharge tube had to be conditioned however by pumping on the copper chloride over night to remove the water vapor and then producing a conditioning plasma which melted the copper chloride powder and deposited a thin film on the walls of the discharge tube. After this conditioning process, a few grams of copper chloride allowed for at least a few hours of continuous operation.

The conclusions of the CW experiments are as follows: CW microwave power of 50 to 100 Watts can vaporize and dissociate copper chloride and produce a uniform copper discharge. By comparing the electric probe measurements of the fields of the cavity to a model of the electromagnetics, we found that this system allows energy

deposition through distances many times the ordinary skin depth (~ 100 cm) as well as achievement of electron densities which are orders of magnitude larger than the critical density ($10^{11} - 10^{12}$ #/cm³). From spectroscopic diagnostics (see the Ph.D. thesis of Will McColl entitled, "Continuous Wave and Pulsed Microwave Resonant Cavity Produced Discharges", Univ. of MI 1993) we concluded that the discharge was dominated by neutral-neutral collisions. Since the copper laser system is a three level system, we knew that we needed to introduce pulsed power to this system to achieve a population inversion.

During the second year we showed that it is possible to *simultaneously* heat and vaporize the copper chloride to the atomic copper states using CW microwaves and then through the use of pulses of microwaves, excite the specific laser states. This combination was achieved by modifying the microwave circuit so the steady state (0-300 Watts) and pulsed (0.5 - 5 kHz, 1 - 4 microsecond pulse length, 0 - 5 kW) microwaves could simultaneous enter the microwave cavity. A coaxial hybrid junction was used to mix these two signals. See Figure 2 for the microwave circuit. As far as we know this was the first attempt to superimpose CW and pulse microwave signals in a microwave discharge. The pulsed microwave signal was found to greatly increase the optical emission of the laser states during the pulse. However, this system also was found to increase the light emission of the non lasing states as well and a population inversion was not achieved. We speculate that this is due to the fact that the lower laser states are metastable and are depopulated through collisions with the discharge tube walls. In this system the metastable states appear to be continually populated and are not allowed to relax. We did try using a smaller discharge tube (13 mm) but the results were similar to the 25 mm tube. We also concluded that the power density was too low to produce the required populations of the upper laser state. See Table I for a comparison of operating conditions for this experiment compared to other configurations including a commercial copper laser.

The above work resulted in a publication in the Rev. of Scientific Instruments, 63(2), Feb. 1992, and is entitled "A combined continuous-wave and pulsed microwave copper chloride discharge". A copy is included in the appendix. The results of these experiments were presented at the 1992 SPIE Intense Laser Beams conference and this presentation was published in the conference proceedings, Vol. 1628. See appendix for a copy.

Gain measurements of the above hybrid system were made using a commercial copper vapor laser. The results were inconclusive because the commercial laser produced varying output powers per pulse. We also attempted resonant shadowgraphy where the spatial absorption profile of the copper discharge is determined by illumination with the commercial laser. Due to non-uniformities in the commercial laser signal, the results from this experiment were also inconclusive. The use of a CCD camera would have made this diagnostic possible, however we do not have such a detector.

We also tried a diagnostic known as laser deflection. In this diagnostic a HeNe laser beam is passed through the discharge and the deflection due to density gradients is measured with a sensitive position detector. The electron density gradients were too small to detect. The thermal density gradients were detectable on long time scales (minutes). Due to vibrations on shorter time scales it was difficult to obtain quantitative results. With more time, we might have been able to overcome the above mentioned difficulties in the gain measurements and the laser deflection diagnostics.

During the third year we attempted to increase the power density to levels used by commercial lasers, See Table I. We purchased a high power (100 kW) magnetron to be used as the microwave source. We had not initially anticipated using a magnetron system and there were no funds available to purchase the needed power supply (~\$100,000). We had at our disposal a used Stanford High Voltage Modulator (30 kV, 40 A, 1 -5 microseconds) which should have been able to drive the magnetron but after two magnetrons and many failed attempts we were not able to generate any microwave

power. Thus we were not able to demonstrate lasing of copper using microwave pumping during the course of this project. However we feel that additional studies at high powers would be very promising.

In an attempt to understand the basic physics of these microwave discharges, we developed additional diagnostics as well as extensive modeling. A double Langmuir probe was designed so that it could be used inside the cavity itself. To the best of our knowledge this is the first application of probes to the interior of the cavity. This work resulted in a publication entitled "A Double Langmuir Probe Diagnostic of a Resonance Cavity Microwave Discharge" and will be published next month (August 1993) in J. Vacuum Science and Technology A. A preprint is attached. We have also improved the theory used to analyze the change in resonant conditions of the cavity due to the discharge (known as the cavity perturbation technique) as well as developed a detailed model of the electromagnetics of the cavity. This work has resulted in a journal article entitled "Electron density and collision frequency of microwave resonant cavity produced discharges", which will be published in the September issue of the Journal of Applied Physics. A preprint is attached in the appendix.

C. ELECTRON BEAM PROGRAM

The electron beam experiments used a Febetron with long - pulse modules which operated at parameters: electron beam voltage of -0.4 MV, electron beam current of ~1 kA and a pulse length of 0.3 microseconds fullwidth. The electron beam was injected through a titanium foil into a vacuum chamber where mixtures of copper vapor resided. We tried three different systems for producing the dense copper vapor needed to make an e-beam pumped laser.

The first system was an external oven designed to generate and transport copper vapor to the electron beam interaction chamber. This system was found to have the problem that the copper chloride condensed on the walls of the quartz tube immediately

upon exiting the oven. This resulted in low copper vapor densities in the e-beam interaction chamber.

In the second system, heat tapes were installed directly on the stainless steel vacuum chamber in order to maximize the density of the copper chloride vapor generated from powder. In order to implement this system it was necessary to install water cooling lines on the vacuum o-ring flange where the interaction chamber connected to the Febetron accelerator. Although this system achieved a temperature greater than 400 degrees C, it took several hours to heat and stabilize and required 3 heat tapes which produced unacceptable heating of the experiment. No significant light emission from atomic copper was detected in e-beam injection experiments with copper chloride in buffer gases of helium and argon.

In the third system, we used heat tapes internal to the stainless steel chamber, wrapped around a Pyrex pipe. This had the advantage that the stainless steel chamber reflected the heat back into the chamber. The system could be heated to required temperatures (>400 C) in a relatively short time. These experiments showed bright He emission lines, due to the helium buffer gas, and possibly some molecular copper chloride emission, but no atomic copper line emission. We believe that the heated copper chloride vapor density was significant, however, the e-beam itself does not dissociate the copper chloride molecules.

In conclusion we feel that a repetitively pulsed electron beam generator is probably needed to both dissociate the CuCl and excite the atomic copper to the laser state.

D. References

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Figure 1. A schematic of the discharge tube used to confine the buffer gas and copper vapor.

Figure 2. Schematic of microwave circuit used in the combined CW and pulsed microwave discharge experiment.

TOP VIEW

Figure 3. Electron beam pumped copper vapor experimental system.

Laser Discharge Conditions				
	Oxford Cu Laser	CW Microwave Exp.	Hybrid Microwave Exp.	Predicted High Power Exp.
Peak input power	50 kW	none	1 kW	150 kW
Time average input power	2 kW	150 W	150 W	300 W
Duty cycle	0.04	continuous	0.003	0.002
Pulse repetition frequency	8 to 14 kHz	none	1 kHz	to 20 kHz
Discharge diameter	25 mm	25 mm or 13 mm	25 mm or 13 mm	25 mm or 13 mm
Discharge length	750 mm	144 mm	144 mm	144 mm
Discharge volume	368 cm ³	71 or 16 cm ³	71 or 16 cm ³	71 or 16 cm ³
Peak power density	135.9 W/cm ³	none	14.1 or 61.4 W/cm ³	2100 or 9200 W/cm ³
Time average power density	5.4 W/cm ³	2.12 or 9.20 W/cm ³	2.12 or 9.20 W/cm ³	4.2 or 18.4 W/cm ³

Table I. A comparison of operating conditions for experimental configurations used in the project and a commercial laser.

E. Publications from this Project

Journal Articles (attached in appendix)

1. "Electron Density and Collisional Frequency of Microwave Resonant Cavity Produced Discharges", W. McColl, C. Brooks, M. L. Brake, to be published in J. Appl. Phys., Sept. 1993.
2. "A Double Langmuir Probe Diagnostic of a Resonant Cavity Microwave Discharge", W. McColl, C. Brooks, M. L. Brake, to be published in J. Vac. Sc. and Tech. A, Aug. 1993.
3. "A Combined CW and Pulsed Microwave Copper Chloride Discharge", W. McColl, M. Passow, and M. L. Brake, Rev. of Sci. Inst. 63(2), 1792, 1992.
4. "Microwave Resonant Cavity Produced Air Discharges", M. Passow, M. Brake, P. Lopez, W. McColl and T. Repetti, IEEE Trans. on Pla. Sci., 19, 219, 1991.

Conference Paper (published proceedings)

1. "Novel Methods of Copper Vapor Laser Excitation", W. McColl, H. Ching, R. Bosch, M. Brake and R. Gilgenbach, SPIE Vol. 1628 Intense Laser Beams (1992), p. 22-31.

Conference Papers (published abstracts)

1. "Perturbation Diagnostics of Microwave Resonant Cavities", W. McColl, C. Brooks, M. Brake, presented to the IEEE Conf. on Plasma Sci, Vancouver, B.C. June 1993.
2. "A Double Langmuir Probe Diagnostic of a Resonant Cavity Microwave Discharge", W. McColl, C. Brooks, M. L. Brake, presented at the 39th American Vacuum Society Symposium, Chicago, IL, Nov. 1992.
3. Mixing of CW and Pulsed Microwave in a Resonant Cavity", W. McColl, M. Brake and M. Passow, presented to the 19th IEEE Conf. on Plasma Sci., Williamsburg, VA, June 1991.
4. "Novel Methods of Copper Vapor Laser Excitation", W. McColl, H. Ching, R. Bosch, M. Brake and R. Gilgenbach, presented to the APS Plasma Physics Conf., Cincinnati OH, Nov. 1990.

5. "Radial and Axial Optical Emission Profiles in Microwave Discharge", M. Passow, W. McColl and M. Brake, presented to the APS Plasma Physics Conf., Cincinnati, OH, Nov. 1990.

APPENDIX

"A Combined CW and Pulsed Microwave Copper Chloride Discharge"

"A Double Langmuir Probe Diagnostic of a Resonant Cavity Microwave
Discharge"

"Microwave Resonant Cavity Produced Air Discharges"

"Novel Methods of Copper Vapor Laser Excitation"

"Electron Density and Collisional Frequency of
Microwave Resonant Cavity Produced Discharges"

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