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Innovative Lasers for Uranium Isotope Separation

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Innovative Lasers for Isotope Separation

Abstract

Copper vapor lasers have important applications to uranium atomic vapor laser isotope separation (AVLIS). We have spent the first two years of our project investigating 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. During the first year, the experiments have been designed and constructed and initial data has been taken. During the second year these experiments have been diagnosed. Highlights of some of the second year results as well as plans for the future include the following:

Microwave resonant cavity produced copper vapor plasmas at 2.45 GHz, have been investigated. A CW (0-500 W) signal heats and vaporizes the copper chloride to provide the atomic copper vapor. A pulsed (5 kW, 0.5 - 5kHz) signal is added to the incoming CW signal via a hybrid mixer to excite the copper states to the laser levels. An enhancement of the visible radiation has been observed during the pulsed portion of the signal. Electrical probe measurements have been implemented on the system to verify the results of the electromagnetic model formulated last year . Laser gain measurements have been initiated with the use of a commercial copper vapor laser. Measurements of the spatial profile of the emission are also currently being made. We plan to increase the amount of pulsed microwave power to the system by implementing a high power magnetron. A laser cavity will be designed and added to this system.

An electron beam pumped copper vapor system (voltage 350 kV, current - 1.0 kA, pulselength 300 ns) has been designed, fabricated and tested. Three copper chloride heating systems have been tested; an external chamber, and externally heated chamber, and an internally heated chamber. Time resolved light emission was observed with filters-PM tubes

and with an optical spectrograph coupled to an optical multichannel analyzer. Molecular copper chloride emission along with helium lines were observed. We plan to increase the density of atomic copper vapor by a combination of a pulsed discharge and e-beam injection system.

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I. 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 two years (9/1/89 - 5/31/91) 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 first innovative laser excitation system we are in the process of investigating uses microwaves to heat and excite lasing transitions in copper vapor plasmas. We have been investigating microwave excitation/pumping systems which utilize an Asmussen microwave resonant cavity. During this first year, we have shown 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). During the second year we showed that it is possible to simultaneously heat and vaporize the copper chloride to the atomic copper states and then through the use of pulses of microwaves, excite the specific laser states. This additional pumping appears to enhance the population of the laser excited states by at least a factor of three.

There are other 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. The most significant advantage of microwave excitation/pumping is that large volumes of plasmas can be generated and sustained. For example, plasma volumes greater than 1000 cm^3 at 2.45 GHz and 2.5 kW have been produced by Bosisio et al.³ We have determined the skin depth to be on the order of 100 cm, indicating the scalability to large radii.

The only previous studies of microwave pumped lasers have centered around rare-gas halogen excimer lasers⁴⁻⁸ and ArXe excimers⁹ where the microwaves were typically coupled to the gas in a waveguide. Lasing in HeNe¹⁰ and argon¹¹ have been demonstrated in France on a small resonant cavity called a surfatron. In general, overall efficiencies of microwave-to-light conversion varied from 0.1% to 1%, comparable to efficiencies achieved with conventional discharges. Some of the advantages to microwave discharge pumped lasers were high microwave deposition efficiency as well as the potential for very high repetition rates and improved control over the laser pulse shape.⁷

The second method we are investigating is electron beam excited/pumped copper vapor lasers. Electron beam excitation/pumping of gas lasers has been demonstrated to be extremely efficient in a number of large-volume gas laser systems¹²⁻¹³, (e.g. CO₂ and KrF). To the best of our knowledge there are no open publications of the investigations of electron beam excited copper discharges with the exception of a recent Russian paper¹⁴ presenting a theoretical model which calculates various characteristics of an electron beam pumped copper vapor laser.

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,¹⁴ 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.¹⁴ estimate that 3% of the electron beam energy could result in laser energy from an electron beam pumped copper vapor laser.

II. Objectives of Research

The overall objects of our three year project are to investigate two innovative excitation/pumping techniques for copper vapor lasers used in uranium isotope separation: 1) Microwaves, and 2) Electron beams. The goals of the research are:

- 1) To demonstrate the feasibility and the advantages of microwave and electron beam excitation/pumping of copper vapor lasers for atomic vapor laser isotope separation:
 - a) Excitation of large copper vapor laser volumes,
 - b) Efficient transfer of microwave and electron beam energy to optical energy of copper vapor lasers,
 - c) Uniform optical laser beam profiles, and
- 2) To develop the scaling laws for application of microwave and electron beam pumped copper vapor lasers to large scale uranium atomic vapor laser isotope separation.

During the first two years we have made progress on achieving part 1a and 1b of the objectives. These results are described in the next section

III. Progress during 9/1/90 - 5/31/91

III. A Introduction

During the first year of our project, the microwave experimental apparatus and the electron beam experimental apparatus were designed, assembled and tested. This past year has centered around diagnosing these systems and trying to optimize their performance. The details of the

microwave pumped copper vapor plasmas are given in Section III B and the details of the results of the electron beam pumped copper plasmas are given in Section III C.

As proposed for this project, we have also purchased a commercial copper vapor laser for amplification studies. Last year this laser was installed in our lab and tested. We have recently begun amplification measurements on the microwave pumped system and we plan to investigate amplification of the electron beam pumped system during the next year.

As mentioned last year, we initially produced atomic copper by heating copper chloride in a quartz tube surrounded by glass wool. The temperature of this oven was controlled by heat tapes powered by a variac. This method turned out to have disadvantages for both excitation/pumping systems. We found that the application of steady state microwaves at about 100 Watts was capable of producing relatively stable and uniform copper vapor discharges, without the aid of the heater tape. This prompted us to use the CW microwaves to heat the copper chloride and then a pulsed microwave signal to produce the laser state excitation. To the best of our knowledge, no-one has attempted to mix separate CW and pulsed microwave signals together in a resonant cavity.

The heat tape system was not optimal for the electron beam pumped discharge system either. There were large temperature gradients, making it difficult to transport the copper chloride vapor to the electron beam deposition area. Two alternative systems to heat copper chloride were designed built and tested. The first involved externally heating the vacuum chamber to temperatures necessary to dissociate the copper chloride. This turned out to be very inefficient and it was difficult to retain adequate vacuum seals between the high temperature portion of the experiment and the rest of the accelerator. The second design uses an internally heated scheme. A recent implementation uses this internally heated system to vaporize the copper chloride and then a high voltage pulse to dissociate the copper chloride. Design of this experiment is given in Section III C.

III. B Progress in the Microwave Pumped Copper Vapor

The experimental system was assembled and tested and preliminary data was gathered. The experiment consists of a microwave circuit for transmitting and measuring 2.45 GHz microwave

power both pulsed and CW signals (see Figure 1), a right angle cylindrical resonant cavity for transferring the microwave power to the laser gas discharge and a quartz tube for containing the gas mixtures. (See Ref. 15 - 18 for cavity details). 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 ~ 2000 K) was surrounded by glass wool in an attempt to keep the wall temperature close to the discharge temperature and to reduce the temperature gradients.

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 cm¹⁸. 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 excite 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 TM_{012} mode.

Steady state (0 - 300 Watts) and pulsed (0.5 - 5 kHz, 1 - 4 microsecond pulse length, 0 - 5 kWatts) microwaves entered the laser cavity through a directional coupler so that incident and reflected power could be monitored simultaneously with calibrated microwave diodes and/or power meters. See Figure 2 for a diagram of this superposition technique. The attenuation of the microwave circuit between the power meters and the coupling probe was calibrated using a network analyzer so that the power delivered to the cavity could be determined. The pulsed and CW signals are mixed in a coaxial hybrid junction. This was found to work very well, however, the main disadvantage to this device is that it splits both signals into two equal portions. Thus half of the total power is currently wasted. Also the used of several connectors also reduces the overall efficiency. This problem will be addresses this coming year by replacement of lossy componenets. We also plan to implement a high power magnetron, to be purchased.

The reactor flow system consists of a mechanical pump, gas bottles of high purity buffer gases (helium), a back pressure regulator, a back pressure monitor and an upstream pressure

Microwave Circuit Diagram

Figure 1. Schematic of cw-pulsed microwave pumped chopper-chloride experiment.

- CW microwaves heat and dissociate copper-chloride
- high power microwave pulse pumps upper laser level

Figure 2. A schematic of the superposition to the CW and pulsed microwave signals.

monitor. As mentioned earlier, heat tape wound around the portion of the glass tube right outside of the cavity was initially used to produce copper vapor from heated copper chloride. About 50 to 100 Watts of CW microwave power heats and vaporizes the copper chloride and the pulsed microwave signals excited the copper atoms.

Optical emission spectroscopy was taken with a 1 meter spectrograph and an image intensified, 1024 element, detector head coupled to an optical multichannel analyzer (OMA). Light produced by the plasma was collected down the axis of the tube for gates of varying durations. There are several conclusions which can be observed from Figures 3 - 6. Figure 3a shows an emission spectrum during the CW portion of the microwave signal only. As can be seen, atomic copper lines prevail indicating that the CW portion of the microwave signal does indeed heat, vaporize and dissociate the copper chloride. Figure 3b shows the spectrum during the peak of the microwave pulse. Note that the copper lines are enhanced, in particular the 5105 Å line which originates from the upper laser state. Figure 4a is an ultraviolet spectrum during the CW portion of the microwave signal and Figure 4b shows a similar spectrum during the pulsed portion of the microwaves. Note similar results to Figures 3a and 3b. Figure 5 shows emission spectra of the visible lines as a function of time. Each data point represents a spectrum taken with 100 nanosecond gates and delayed relative to the start of the microwave pulse. Note that the 5105 Å line follows the microwave pulse and shows a dramatic increase during the microwave pulse. A similar increase is seen for the 3274 Å ultra-violet lines, see Figure 6.

Note that all of our studies to date have centered around helium at pressures between 1 and 35 Torr. We plan to investigate the effects of other buffer gases, most notably that of neon.

Last year we developed a comprehensive model of the electromagnetics of the system,¹⁹ (See Appendix I). Currently we are developing a probe diagnostic to try and determine the plasma temperature and density so that the results of this model can be checked.

Figure 3a. A visible emission spectrum taken with a 100 ns gate during the CW portion of the microwave signal. The plasma conditions were 2Torr helium, 101 Watts CW power.

Figure 3b. A visible emission spectrum taken with a 100 ns gate during the peak portion of the pulsed + CW microwave signal. The plasma conditions were 2 Torr helium, 101Watts CW power and 90 Watts of pulsed power at a rep rate of 500 Hz and pulse length of 4 μ s.

Figure 4a. An ultra-violet emission spectrum taken with an 100 ns gate during the CW portion of the microwave signal. The plasma conditions were 2 Torr helium, 101 Watts CW power.

Figure 4b. An ultra-violet emission spectrum taken with an 100 ns gate during the peak portion of the pulsed + CW microwave signal. The plasma conditions were 2 Torr helium, 101 Watts CW power and 250 Watts of pulsed power at a rep rate of 500 Hz and pulse length of 4 μ s.

Figure 5. The visible emission lines as a function of time. Each data point represents a spectrum taken with 100 nanosecond gates and delayed relative to the start of the microwave pulse.

Figure 6. The ultra-violet emission lines as a function of time.

We also plan to investigate the spatial uniformity of the plasma. This will be performed by examining the radial light emission with the spectrograph and OMA where in this case the detector array will be rotated by 90° at the exit plane of the spectrograph. Thus the detector will measure light at one wavelength as a function of position. The spatial uniformity will be optimized as a function of power level, pulse repetition rate, pulse length, type of buffer gas and pressure.

Laser amplification measurements have been attempted with the commercial copper laser bought with this project. Amplification has not been demonstrated, however some of the results are ambiguous and require further investigation. For example, refractions in the copper vapor may have to be accounted for. As mentioned earlier, we are attempting to increase the amount of power delivered to the copper vapor. The power density effects on amplification will be studied during this next year.

Also during the next year, we plan to design and implement a laser cavity. The effects of the various input parameters such as power, pulse repetition rate, pulse duration, gas pressure etc. on laser light output will be examined.

III. C Progress on the Electron Beam Pumped Copper Vapor Laser Experiment

During the second year of this research program we have experimentally investigated several techniques for producing dense copper vapor, injecting electron beams into these mixtures and diagnosing the interaction. Recall that in last year's report we described System I, an external oven which was 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 order to overcome these problems we developed three new experimental configurations to generate dense copper vapor in an e-beam interaction chamber:

- 1) Externally heated stainless steel chamber, System II (Figure 7),
- 2) Heated pyrex chamber internal to the stainless steel cross, System III (Figure 8), and

3) Electrical discharge system located inside the heated pyrex chamber internal to the stainless steel cross, System IV, (Figure 9).

Each of these systems will be discussed below.

The experimental configuration of System II is depicted in Figure 7. A Febetron electron beam generator (300 kV, 1 kA, 300 ns) is coupled to the copper vapor laser chamber through a screen and a 1 mil titanium foil with an extracted current between 200 A to 400 A. Heat tapes were installed directly on the stainless steel chamber in order to maximize the density of the copper chloride vapor generated from powder. Thermocouples were used to verify that the temperature of the cross was greater than 400 degrees C. 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.

Experimental System III (Figure 8) 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. Electron beam injection experiments were performed on system III with helium and argon buffer gas. These experiments showed bright He emission lines 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.

The latest version of the experiment, System IV, was fabricated to alleviate the problems of the previous 3 versions. This experimental configuration, depicted in Figure 9, had discharge electrodes internal to the heated quartz tube. The idea here is that the discharge could dissociate the copper chloride molecules so that the e-beam could be injected into atomic copper vapor.

TOP VIEW

Electron-beam pumped copper vapor experimental system II

TOP VIEW

Electron-beam pumped copper vapor
experimental system III

**Electron-beam pumped copper vapor
experimental setup IV (with discharge system)**

Experimental System IV has been built and tested. A single experimental run has been conducted; however, the results were inconclusive due to vacuum leaks. This experimental system will be the starting point for the next year's research program.

IV. Proposed Research Program

IV. A Introduction

As mentioned, the research program consists of two major areas which are being investigated in parallel:

- 1) Microwave excitation/pumping of copper vapor lasers; Prof. Brake and one graduate student will concentrate on these investigations, and
- 2) Electron beam excitation/pumping of copper vapor lasers; Prof. Gilgenbach and one graduate student will concentrate on this area.

The research objectives and optical diagnostics are similar for both the microwave and electron beam excitation however, as expected, the experimental design of the two experiments is quite different. Two separate copper vapor laser chambers are being utilized for the microwave excitation experiments and the electron beam excitation experiments. However, optical diagnostic hardware, (e.g. spectrograph, optical multichannel analyzer, dye laser etc.) are shared between both thrust areas. Experimental techniques specific to microwave or electron beam excitation are discussed separately in the following sections.

The experiments will focus on the following tasks over the final year of the project:

- 1) Optimize copper vapor discharge parameters as a function of input parameters such as delivered microwave and electron beam energy or power, discharge pulse-length, pulse repetition rate, and buffer gas pressure:
 - copper line emissions using emission spectroscopy
 - amplification using the commercial copper vapor laser
 - radial uniformity of excitation/pumping
 - overall efficiency

2) Add a laser cavity (Brewster angle windows and laser mirrors) and investigate lasing of the copper discharges. We plan to measure the following important copper vapor laser parameters as a function of time while varying input parameters such as delivered microwave and electron beam energy or power, discharge pulse-length, pulse repetition rate, and buffer gas pressure:

- laser gain
- laser power
- overall efficiency

3) During the later part of the third year we will evaluate the scaling laws for microwave pumped and electron beam excited discharge lasers for large scale copper vapor laser systems, and

4) Assess the impact of these microwave and electron beam laser excitation/pumping systems on the technology of large scale atomic vapor laser isotope separation.

IV. B Proposed Research for Microwave Pumping of Copper Vapor Lasers

Experiments on microwave pumped copper vapor will continue to concentrate on research to generate copper discharges with suitable parameters for copper vapor lasers. We plan to increase the total microwave power delivered, particularly during the pulsed portion of the operation.

Microwave excitation of the 510 nm and the 578 nm lines will be optimized as a function of

- 1) microwave pulse repetition rate
- 2) microwave pulse length
- 3) the performance of helium versus neon buffer gas.

Also plasma uniformity and external laser amplification will be optimized as a function of input parameters. Additional diagnostics such as Langmuir probes will be added to the system to verify the model developed last year.

A laser cavity consisting of Brewster angle windows attached to the discharge tube and highly reflecting mirrors, placed outside of the tube, will be added to the system. The presence of stimulated emission will be measured with calibrated photo diodes and a calorimeter. This output will be optimized as a function of input parameters such as power, pressure and pulselength.

IV. C Proposed Research for Electron Beam Pumping of Copper Vapor Lasers

Proposed research during the third year of this research program will concentrate on the generation and pumping of copper vapor by long-pulse relativistic electron beams. The initial experimental configuration will be the internally-heated pyrex tube with auxiliary discharge, System IV, shown in Figure 9. Experiments will be performed by heating the copper vapor and adjusting the delay between the auxiliary discharge and the electron beam. Initially, the delay will be such that the discharge fires first, in order to dissociate the copper vapor prior to electron beam injection. The relative timing of the discharge-e-beam delay will be optimized to produce the most intense spectroscopic emission from atomic copper lines.

Several different buffer gases will be investigated for these experiments: helium, neon, and argon. The effect of the different stopping power of these gases for electron beams will be explored. Transported e-beam current can be compared to injected current in order to estimate the e-beam energy deposition in the buffer gases.

A new experiment which will be investigated is a hybrid microwave- generation /e-beam- excitation copper vapor system. This configuration, System V, is depicted in Figure 10. This will combine the advantages of microwaves for generation of a dense atomic vapor and the advantages of electron beams for intense high-power excitation of the resulting vapor.

After the optimal electron-beam-excited atomic copper emission condition has been established for these experiments, a series of small-signal gain experiments will be performed on the most promising configuration. These experiments will utilize an existing commercial Oxford Lasers copper vapor laser. Upon demonstrating small-signal amplification, laser cavity mirrors can be added to investigate oscillation of e-beam-excited copper vapor.

Electron-beam pumped copper vapor experimental system V (microwave e-beam hybrid)

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