OUTLOOK FOR METAL LASERS*

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Lasers using atomic metallic vapors have drawn considerable attention since their invention in 1965 because of their potential for producing high power in the visible spectrum. Because, as yet, none of these lasers can be operated continuous wave (CW), the potential of high average laser power is tied to the attainment of both high laser energy density per pulse and the achievement of large pulsing rates.

Early attempts to attain lasing in metallic vapors utilized single pulsing in heated discharge tubes (refs. 1 and 2). Although lasing was demonstrated in a number of different metals, copper vapor was found to have the largest energy output per pulse and the best efficiency. Following the earlier work in heated discharge tubes, flowing laser devices were constructed to achieve multiple pulsing where the pulsing rate would not be limited by the laser kinetics, but only by the lasant flow velocity through the optical cavity (refs. 3 and 4).

Many of the early attempts to demonstrate lasing in both static and flowing laser systems were successful. However, the potential of this class of cyclic laser remained in doubt because of the high operating temperatures they required and the attendant and serious materials problems and heat transfer losses associated with vaporizing the metals. As an example, static copper vapor lasers generally operate at about 1500°C, and the stagnation temperature of supersonic copper vapor lasers can exceed 2000°C.

It has been apparent for some time to the workers in this field that, if metallic compounds having lower melting points than pure metals and correspondingly high vapor pressures at a given temperature could be utilized, the high operating temperatures could be lowered appreciably. Attempts at JPL and other laboratories to use metallic halides and organic metallic compounds with single discharges have heretofore failed to produce any power output from a laser cavity. Recent work carried out at JPL (ref. 5) using copper chloride and other metallic halides has indicated that this failure occurs because it is not possible to obtain simultaneous dissociation and lasing in a single electrical discharge. The dissociated metal from a single discharge contains an appreciable population of the lower metastable lasing levels which prevents lasing.

A double-discharge technique developed at JPL has proven to be a way of circumventing this problem. The metallic compound is heated and vaporized in a laser cavity and then is dissociated by an electrical discharge. A population inversion is attained in a subsequent second discharge applied at an appropriate time delay after the first discharge. There is an optimum time interval between the two discharges and an optimum lasant temperature for maximum laser power output per laser pulse.

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To date, lasing has been attained utilizing the double-discharge technique in manganese chloride, lead chloride, copper chloride, copper iodide, and copper formate (refs. 6 through 7).

Characteristics of these lasers are summarized in table 1. Although lasing can be attained in copper formate in several sequential double discharges, decomposition of the copper formate subsequently causes carbon plating on the laser windows terminating the laser power output. Therefore, no reliable energy or power density data could be obtained using this compound. Because copper chloride appears to be the best lasant investigated thus far, a comprehensive parametric and spectroscopic absorption study has been initiated at JPL to determine the most dominant features of the laser kinetics and to optimize the efficiency and power output of both static and flowing copper chloride laser devices.

In heated copper chloride laser discharge tubes the power output is a function of the time delay between electrical discharges, the lasant temperature, the tube length and diameter, buffer gas composition and density, the optical cavity characteristics, and the power supply and laser electrical circuit characteristics.

Typical data for a copper chloride lasant, demonstrating the effect of time delay after the first electrical discharge and the effect of lasant temperature, are shown in figure 1. For times shorter than the optimum delay time the lower metastable levels of the copper are not depleted enough to establish a large inversion. For times larger than the optimum, chemical recombination of the copper and chlorine causes a decrease in the excitation of the upper lasing levels because of a diminishing of the copper ground state population. A diagnostic study utilizing absorption techniques is being conducted to measure the temporal variation of the copper lower metastable states and the ground state. The copper ground state density is determined by the absorption of radiation from a pulsed xenon flashtube at 3247 Å, the wavelength of one of the copper resonance lines. One of the metastable levels, the lower level of the lasing transition at 5106 Å, is determined by absorption at 5106 Å. The results obtained thus far confirm the original hypothesis that the copper metastable population is large following the first electrical discharge, and that these metastable levels must decay appreciably before lasing can be achieved in a second pulse.

Figure 2 shows the results of some absorption measurements utilizing a 13 mm diameter laser tube. Three types of measurements are indicated in the figures. The three upper curves are obtained from absorption measurements at the resonance wavelength of 3247 Å at three different lasant temperatures. The lowest of these three curves was obtained at a temperature just lower than the lasing threshold. A single curve at the lower left resulted from absorption of the 5106 Å line. For these measurements, only the dissociation pulse was activated and the abscissa represents the time delay between the dissociation pulse and the peak of the xenon flash.

The remaining two curves, representing the laser energy, were obtained at the same conditions as the absorption measurements but with the laser mirrors in place and the laser pumping pulse operating. For the purpose of displaying the laser energy on a basis that is directly comparable with the absorption measurements, the measured radiation at 5106 Å was converted to the number of copper atoms necessary to produce the measured radiation at 5106 Å. Since a mirror having a transmission of 16 percent was used as the output mirror, the number density indicated represents the minimum number of atoms, in the $^2P_{3/2}$ state, that must be stimulated to produce the measured energy. The chief conclusion one can draw from figure 2, which is representative of most of the data taken thus far, is that the onset of lasing is definitely dependent on the rate of decay of the copper lower metastable levels, and that this rate of decay is much faster than the chemical recombination rate.

In figure 1, a plot of laser power output as a function of lasant temperature at a fixed time delay exhibits an optimum operating temperature of about 400°C. This optimum temperature is also applicable to other laser tube geometries and laser operating conditions. It is understandable that at low temperatures the copper chloride density is too low to provide enough copper atoms for lasing. However, the reason for the decrease in the laser power beyond the optimum temperature at increasing copper chloride densities is not clear at this time. It is this facet of the operation of the copper chloride laser that is currently being intensively studied; particularly to determine if the limitation in increases in the lasant density is of a fundamental nature or whether it is associated with the particular laser system that is currently under study.

Because of the relatively slow chemical recombination relaxation time compared to the time required for the evolution of the laser kinetics, it was decided to pulse continuously a copper chloride-helium mixture where the laser waste heat would be used to vaporize the copper chloride as was demonstrated in pure metal vapors in reference 8. Multiple pulsing was carried out in discharge tube diameters using frequencies corresponding to data taken in double-discharge heated laser tube experiments (fig. 3). Note that the optimum delay time decreases as the discharge tube diameter decreases, indicating that diffusion as well as electronic deexcitation is acting to deplete the lower lasing levels between current pulses.

Multiple lasing is observed when a train of single identical current pulses are spaced according to the data shown in figure 3. The initial current pulse acts as a pure dissociation pulse; successive current pulses serve both as laser pumping and dissociation pulses. Similar results have been obtained recently with copper iodide used as the lasant (ref. 9).

Data have been obtained utilizing multiple pulsing for discharge diameters varying from 1 to 4 cm (ref. 10). Although insufficient data have been obtained to determine the limits on laser efficiency at small pulsing rates, the data obtained thus far show that the efficiency increases as the pulsing rate increases. As the pulsing rate is increased beyond the corresponding optimum rates obtained in the double-discharge experiments, one would expect this trend to reverse because of an insufficient time between pulses required to deplete the lower lasing levels. However, this point has not been reached yet in the current experimental work.

To date, the best data have been obtained at 5106 Å using a 30-cm length and a 1-cm-diameter discharge tube with a pulsing rate of 2×10^4 sec^{-1}. At an average efficiency of 1 percent, the energy and power density per pulse are 35 μJ cm^{-3} and 1.7 kW cm^{-3} respectively; the laser pulse width is 20 nsec and total average power output is 16.5 W at an average power density of 0.7 W cm^{-3}. The efficiency is defined as the ratio of the laser energy output and the energy stored in the capacitor. Thus far, only a helium buffer gas has been used with the copper chloride lasant in the multiply-pulsed experiments. The best laser performance is obtained at 10 torr. The effect of pressure and buffer gas composition will be the subject for future study.

In the multiply-pulsed experiments, although the lasant temperature is initially maintained with a laser heater as in the double-discharge experiments, the heater is turned off when the data are obtained. Without exterior cooling, multiply-pulsed operation can only be continued for a few minutes before the laser waste heat raises the lasant temperature and the performance decreases in a manner similar to that observed in the double-pulsed experiments. The average laser power measured with a thermopile agrees, within experimental error, with the power calculated from the time-averaged values of the pulse power obtained with a photodiode. The laser output is very continuous with no missing laser pulses; a laser pulse is obtained with every current pulse. This feature of a one-to-one correspondence between laser and current pulses is shown in figure 4.

An average laser efficiency of 1 percent can probably be improved with further optimization of the discharge tube geometry and power supply characteristics, but our best estimate is that the improvement will not exceed 10 percent and more probably be of the order of 1 to 3 percent. In order to increase the average power from a copper chloride laser beyond that which can be attained from a static system, which appears to be about 100 W, one is faced with two problems. The first is removal of the waste heat deposited in the optical cavity with each electrical discharge, and the second is the upper limit in the pulse repetition rate imposed by the rate of relaxation of the copper lower lasing levels.

One possible solution to both problems is the technique of flowing the copper vapor through the optical cavity at high velocities. This technique removes the waste heat by forced convection and, at the same time, by replenishing the copper vapor charge in the optical cavity between pulses, allows higher pulse repetition rates and therefore higher average powers. To date, repetition rates of approximately 10^4 Hz have been achieved, for short periods, in static systems. To increase this rate one needs velocities greater than 10^4 cm/sec for a 1-cm-diameter cavity. A velocity of 10^5 cm/sec would increase the average power tenfold.

A system designed to study this technique is shown schematically in figure 5. The essential features of the system are an electrically heated graphite container that is used to continuously vaporize the copper chloride, a plenum chamber into which preheated helium is injected and mixes with the copper chloride vapor, a subsonic electrical discharge channel, and a supersonic nozzle to expand the halium-copper chloride mixture into a supersonic jet. The dissociation pulse is applied in the subsonic flow prior to the supersonic expansion. The second lasing pulse is positioned in the supersonic flow at a distance downstream from the dissociation pulse such that the flow time between the two discharges corresponds to the optimum delay time in a static double-pulsed discharge tube. Studies conducted with this supersonic laser facility have been partially completed. Parameters such as the location and electrical characteristics of the dissociation and pumping current pulses, optical cavity characteristics, copper chloride vapor density, and control of boundary layers have been investigated. Results obtained to date show that the copper chloride vapor remains supersaturated in the supersonic expansion, confirming the theoretical predictions made using non-stationary nucleation theory (ref. 11). Laser power extraction in the supersonic flow has been attained at 5106 Å with laser energy densities of about 2.5 μJ cm^{-3} at flow velocities exceeding 10^5 cm/sec, thus demonstrating that a supersonic copper chloride laser can be pulsed at rates of 10^5 Hz.

Work is continuing to determine what problems would be encountered in a much larger supersonic laser. At this time, there do not appear to be any fundamental problems associated with scaling up this laser device to produce average laser powers in the multi-kilowatt regime. In addition, larger supersonic jets will allow the number of lasing pulses to be large compared to the single initial dissociation pulse, so that the flowing laser has the potential of attaining a 1 percent or better efficiency, equal to the efficiencies currently being attained in multiply-pulsed laser discharge tubes, but with laser pulsing rates an order of magnitude faster.

REFERENCES

1. Piltch, M.; Walter, W. T.; Solimene, N.; Gould, G.; and Bennett, W. R. Jr.: Pulsed Laser Transitions in Manganese Vapor. Appl. Phys. Lett. Vol. 7, no. 11, 1 Dec. 1965, p. 309-310.

2. Walter, W. T.; Solimene, N.; Piltch, M.; and Gould, G.: Efficient Pulsed Gas Discharge Lasers, IEEE J. of Quant. Elect. Vol. QE-2, no. 9, Sept. 1966, p. 474-479.

3. Ferrar, C. M.; Copper Vapor Laser with Closed Cycle Transverse Vapor Flow, IEEE J. Quant. Elect., Vol. QE-9, Aug. 1973, p. 856-857.

4. Russell, G. R.; Nerheim, N. M.; and Pivirotto, T. J.: Supersonic Electrical-Discharge Copper Vapor Laser. Appl. Phys. Lett., Vol. 21, no. 12, 15 Dec. 1972, p. 565-567.

5. Chen, C. J.; Nerheim, N. M.; and Russell, G. R.: Double-Discharge Copper Vapor Laser with Copper Chloride as a Lasant. Appl. Phys. Lett., Vol. 23, no. 9, 1 Nov. 1973, p. 514-515.

6. Chen, C. J.: Manganese Laser Using Manganese Chloride as a Lasant. Appl. Phys. Lett., Vol. 24, no. 10, 15 May 1974, p. 499-500.

7. Chen, C. J.: Lead Laser using Lead Chloride as a Lasant. J. Appl. Phys., Vol. 45, no. 10, Oct. 1974, p. 4663-4664.

8. Isaev, A. A.; Kazaryan, M. A.; and Petrash, G. G.: Effective Pulsed Copper-Vapor Laser with High Average Generation Power. Soviet Physics JETP Lett., Vol. 16, no. 1, July 1972, p. 27-29.

9. Liberman, I.; Babcock, R. V.; Liu, C. S.; George, T. V.; and Weaver, L. A.: High-Repetition-Rate Copper Iodide Laser. Appl. Phys. Lett., Vol. 25, no. 6, 15 Sept. 1974, p. 334-335.

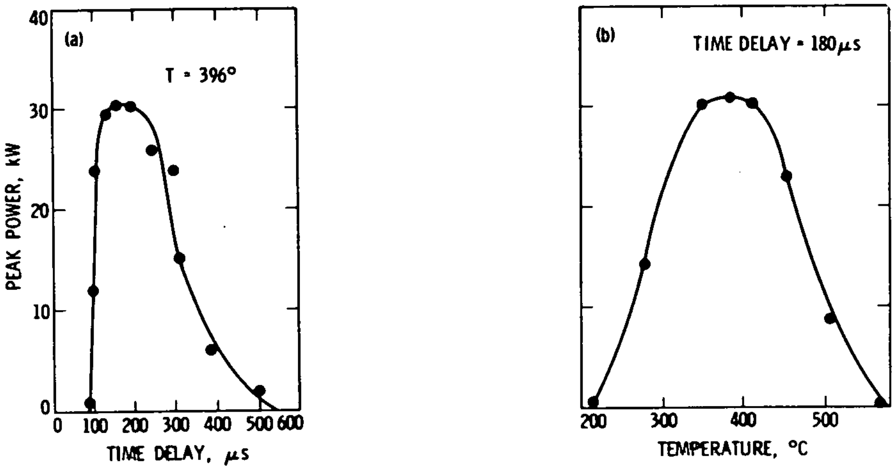
10. Chen, C. J.; and Russell, G. R.: High-Efficiency Multiply Pulsed Copper Vapor Laser Utilizing Copper Chloride as a Lasant. Appl. Phys. Lett., Vol. 26, no. 9, May 1975, p. 504-505.

11. Harstad, K. G.: Nonstationary Homogeneous Nucleation. J. Heat Trans., Vol. 97, no. 1, Feb. 1975, p 142-144.

TABLE 1. – OPERATING CHARACTERISTICS OF DOUBLE PULSED METALLIC VAPOR LASERS

Operating parameters	Lasant				
	Manganese chloride	Lead chloride	Copper chloride	Copper iodide	Copper formate
Time delay[a], μsec	150	150	100	100	100
Temperature, °C	680	500	400	575	135
Buffer gas, torr	He at 1-2	He at 1-2	He and Ar at 1-20	He at 1-2	He at 1-2
Laser energy density, μJ/cm^3	1.3	4	35	11	–
Laser peak power density, W/cm^3	33	160	1700	500	–
Wavelength, Å	5341	7229	5107	5107	5107

[a] 1-in. - diameter tubes



(a) Time delay between pulses.

(b) Temperature.

Figure 1.– Dependence of laser peak power.

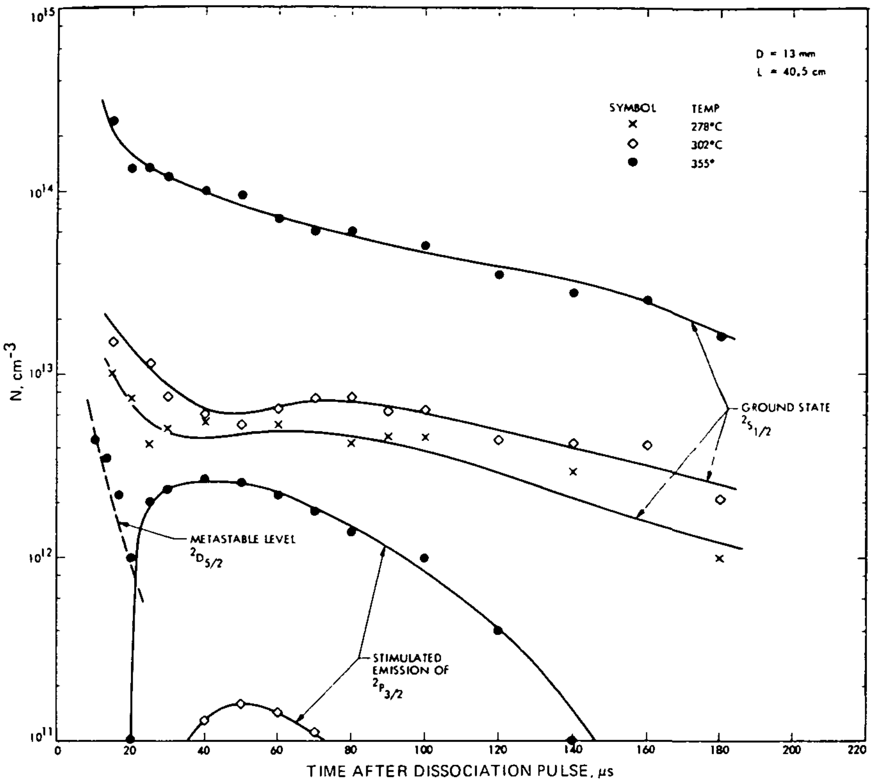


Figure 2.– Measured densities of copper ground state and metastable levels as a function of time after the dissociation pulse. Laser energy at the same lasant conditions is also shown as the minimum number of stimulated emissions needed to produce the measured laser energy.

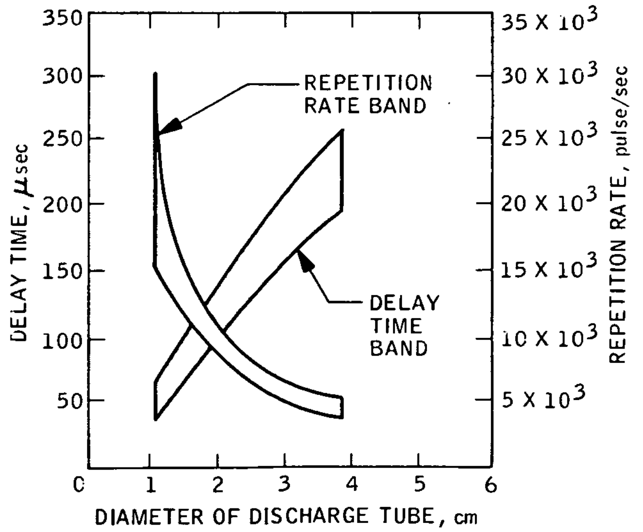


Figure 3.– Delay time and repetition rate as a function of laser tube diameter.

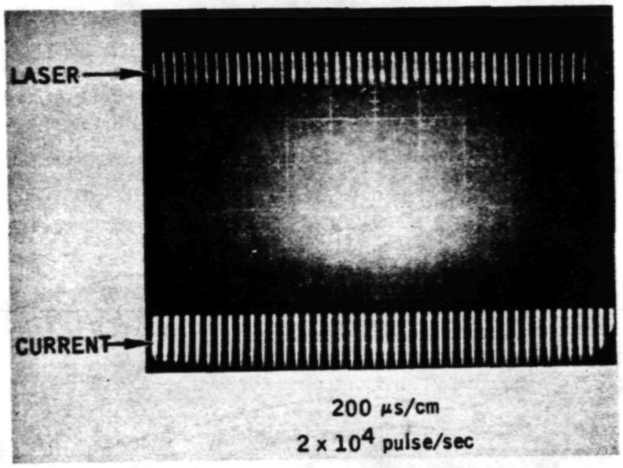


Figure 4.– Oscillogram of current and laser pulses.

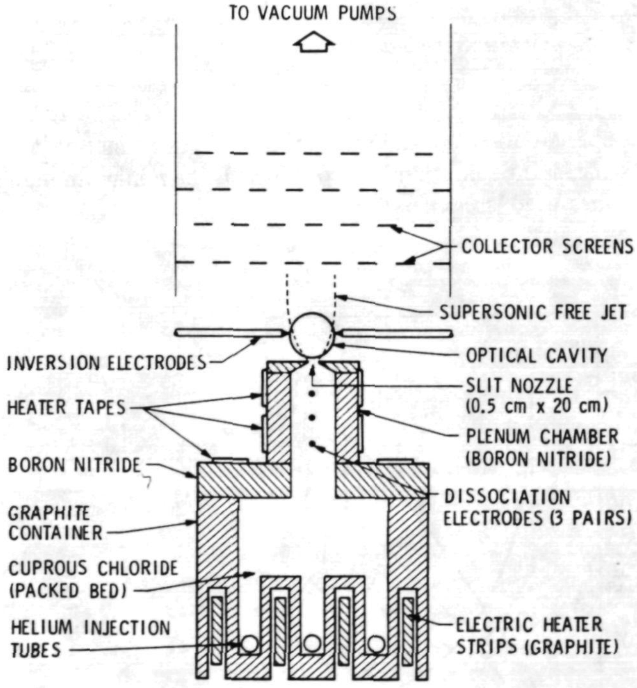


Figure 5.– Schematic of supersonic copper chloride laser.

DISCUSSION

Tom Karras, General Electric Co. – You mentioned an average power of 0.7 W. Was that per unit volume?

Gary Russell: Yes 0.7 W/cm^3.

Tom Karras: Thus the 30 W comes from a 40 cc system?

Gary Russell: Right.

Ned Rasor, Rasor Associates: – Would a pure copper vapor laser be more efficient, that is, does dissociation take much of your power?

Gary Russell: In the double-pulsed experiments we were surprised that, under certain conditions, the dissociation pulse could take up to ten times the laser pulse energy. But when you continuously pulse it, as we are doing here, the chemical recombination time is so much slower that you don't have to start over every time. So you have a very small amount of energy being used for dissociation in each pulse, that is, just enough to make up for the small amount of recombination between pulses.

Tom Karras, General Electric Co. – Just a comment. For our stationary copper vapor system our numbers are very comparable.

Gary Russell: Yes. I should also mention that the chlorine just acts like a buffer gas. Our theory indicates that it influences the discharge a little, but it doesn't enter kinetically in any way.

Ernest Brock, Los Alamos Scientific Lab. – Not to detract from your 1 percent efficiency here, but what do you have in mind for pulsing to higher efficiencies?

Gary Russell: I would say an upper bound in a static system is 10 percent. We are shooting for somewhere between 1 and 3 percent. In the flowing systems, the small systems are going to be inefficient because you can't pulse many times in flow. If you go to a large enough system, however, say a 100 kW system, 1 J per pulse, then the volume would be large enough so that you can discharge many, many times in the flow in addition to the one dissociation pulse. You are then back in the same kinetic situation as you are in the static tubes where the dissociation losses are small compared with the lasing losses. So with the flowing system, when it's made large, and if the aerodynamic problems are not severe, I would say we should approach 1 percent efficiency.

Mark Wrighton, M.I.T. – Have you considered using volatile metal compounds such as tetraethyl lead which can photalyze at room temperatures to give metal atoms?

Gary Russell: No we haven't since the copper chloride and lead chloride work so well. One thing we must keep in mind in the static systems is that you don't necessarily want to go to too low temperatures since you want to conduct waste energy out of the tube.

Abe Hertzberg, University of Washington – Gary, I want to compliment you on a fine piece of work. When I see people struggling with improving the efficiency of their argon ion lasers to the levels you are starting from, it only indicates how much of an accomplishment it is.