Characteristics of the Pumping Pulse and the Output Laser Pulse for a Cu/CuCl Double Pulse Laser

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Abstract–**Characteristics of the pumping discharge pulse and laser pulse in a Cu/CuCl double pulse laser have been measured as a function of time delay, buffer gas pressure, and tube temperature. We have found that for otherwise fixed discharge conditions, pumping rates decrease as these quantities are increased. The shape of the laser pulse as a function of time delay is shown to be dependent on the rate of current rise of the pumping pulse. The length of time required by the pumping pulse to achieve threshold is found to be a function of time delay, buffer gas pressure, and tube temperature. The implications of this behavior for the role of metastable copper and its mode of relaxation are discussed.**

I. Introduction

THE copper laser has received much attention in recent years as a source of intense optical pulses [1]-[5]. Unlike the conventional copper laser which requires a tube temperature of about 1500°C to obtain sufficient copper vapor from pure copper [4], the use of copper chloride (CuCl) as a source enables optimum tube temperatures to be near 400°C [1]. To obtain laser action from CuCl vapor at least two discharge pulses are required. The first discharge pulse dissociates the CuCl, producing copper atoms. The second discharge pulse pumps the copper atoms, producing the laser pulse. The discharge pulses are typically 1-20 nF at 12-20 kV charging voltage.

Because a large fraction of the copper atoms emerge from the dissociation in the metastable lower laser level, there is a minimum time delay between discharge pulses which must pass before enough copper atoms have collisionally relaxed to permit oscillation. Copper atoms are continually reassociating to form the parent molecule, and there is a maximum delay time between discharge pulses beyond which threshold cannot be reached because the copper atom density is too low. Between the minimum delay (a few to tens of microseconds) and the maximum delay (tens to hundreds of microseconds), there is an optimum time delay for which output pulse energy is maximum.

The afterglow period between discharge pulses is important primarily for two reasons. The first is that during this period thermal collisional processes occur (e.g., reassociation, charge exchange, collisional deactivation) which determine the availability of ground-state copper. The second is that initial conditions are provided for the pumping discharge pulse. It is clear that thermal processes cannot significantly affect laser pulse energy during the pumping pulse since the laser pulse is only tens of nanoseconds wide and it appears as early as the first few nanoseconds of the pumping pulse. The complicated dependence of the laser pulse energy on time delay cannot be explained on the basis of the pumping pulse alone. An accounting of the processes occurring during the interpulse afterglow is required.

When all other controllable discharge parameters are fixed, laser pulse energy has been shown to be a function of the current rise time for the pumping pulse. By increasing the rate of current rise the laser pulse energy can be increased [6]. Even though charging voltage, tube temperature, and gas pressure remain constant, the pumping pulse, rate of current rise, and hence pumping rates cannot be expected to remain unchanged as the time delay is varied. For short time delays, the afterglow electron density is relatively high so that the impedence of the discharge tube is low. The pumping pulse does not need to breakdown the gas and the rate of current rise is large. For long delays, the afterglow electron density is low and the density of molecular CuCl is large compared to the values for shorter delays. The impedence of the discharge tube is higher, the rate of current rise is smaller, and average electron energy is lower. Hence, pumping rates (i.e., the rates at which the inversion can be produced) decrease as the time delay increases.

We report in this paper the results of measurements of characteristics for the pumping discharge pulse and the laser pulse as functions of time delay in a Cu/CuCl double pulse laser. It will be shown that the dependence of laser pulse energy on time delay, gas pressure, and tube temperature (CuCl vapor pressure) is partly due to the dependence of the rate of current rise, peak current, and hence pumping rate on these same quantities. An analysis of the laser pulse based on rate equations will be used to show that the dependence of laser pulse energy, power, and the shape of the laser pulse on the pumping rate is closely related to the gain switched behavior exhibited by the copper laser.

The pumping time is the length of time required by the pumping pulse to achieve threshold; it too is a funciton of time delay and pressure. At short delays, the pumping time required is long. This time decreases to some minimum value as the time delay increases, and then increases at longer time delays. The time delay and pressure dependence of this behavior will be related to the mode of relaxation of metastable copper.

II. Experimental Results

The laser discharge tube was made of pyrex and had a 12 mm ID. The pin electrodes were spearated by 65 cm. The middle 60 cm of the tube was externally heated by an electrical heater. Research grade helium buffer gas was passed by the

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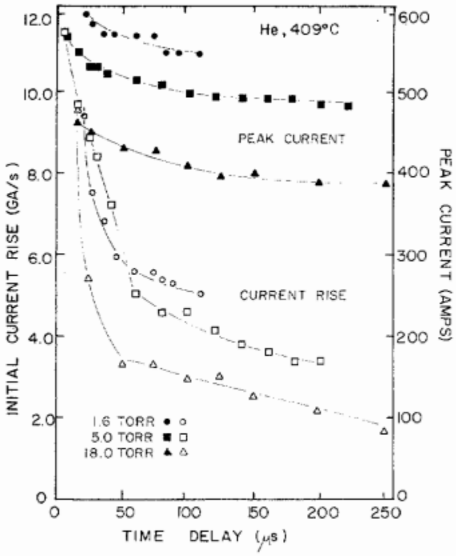

Fig. 1. The rate of current rise and peak current of the pumping pulse as a function of time delay and buffer gas pressure in helium at 409°C (CuCl vapor pressure ≈0.04 torr). Charging voltage was 16.0 kV and discharge capacitance was 5 nF.

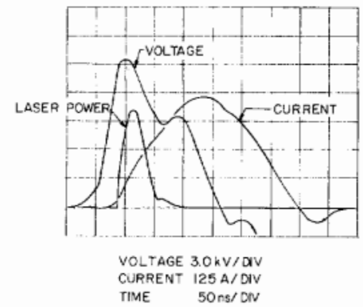

Fig. 2. Typical pumping pulse current and voltage, and laser power. Note the small voltage required to initiate the current. Also note that the laser pulse appears at the leading edge of the current pulse.

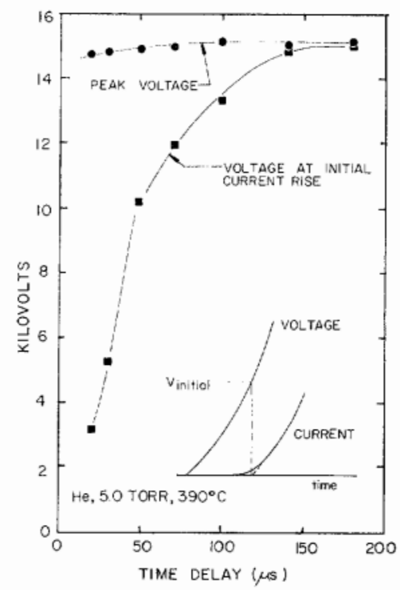

Fig. 3. Peak voltage across the discharge tube and the voltage at which the current begins to rise during the pumping pulse. The voltage at which the current begins to rise is defined by the sketch above.

fused silica windows to prevent CuCl from depositing on the cooler window surfaces. There was no net gas flow through the tube. The double pulse power supply is described in detail elsewhere [6]. The dissociation pulse and pumping pulse capacitors were 5 nF charged to 16 kV. The optical cavity consisted of a 2 m, 99 percent reflecting dielectrically coated mirror and a quartz flat output coupler. Discharge pulse current was measured with a Pearson 411 current transformer and displayed on a Hewlett-Packard 1722B oscilliscope. Discharge voltage was measured with a 1000:1 voltage divider. Laser energy and power were measured with a Korad KD-1 photodiode.

The rate of current rise for the pumping pulse and peak current as a function of time delay and buffer gas pressure are shown in Fig. 1. Note that as the time delay increases, both the rate of current rise and the peak current decrease. For constant time delay, increasing the pressure decreases the rate of current rise. The decrease in these quantities with increasing time delay is likely due in part to the decrease in afterglow electron density and increase in plasma impedence. The decrease with increasing pressure is partly due to dependence on E/N and partly due to the more rapid rate of electron cooling (and hence recombination) experienced at a higher pressure. The dominant energy loss mechanism for electrons during an afterglow is elastic collisions with the buffer gas. In helium at low electron energy, the elastic collision rate is inversely proportional to the electron temperature and proportional to the gas pressure [7]. The dominant form of electron-ion recombination is collisional radiative varying with electron temperature as $T_e^{-9/2}$ [8]. Hence, the higher the gas pressure, the more rapidly the electrons cool, and the more rapidly they recombine. The rate of current rise, which is inversely proportional to the discharge impedence, therefore, decreases with increasing buffer gas pressure and increasing time delay.

The relationship between pumping pulse voltage and the initial current rise as a function of time delay provides further evidence of the importance of initial conditions provided by the afterglow. A typical trace of laser power, pumping pulse current, and voltage is shown in Fig. 2. Note that the current begins to flow before the voltage reaches its peak value. If this were a classical breakdown, the peak voltage and initial current rise would very nearly coincide. The voltage at which the current initially rises is therefore an indication of the impedence of the discharge.

The voltage at which the current initially rises and the peak voltage sustained by the tube as a function of time delay are shown in Fig. 3. We have found similar behavior for a wide range of buffer gas pressures and tube temperatures. Note that as the time delay increases, the voltage required to initiate the pumping pulse current increases. At short time delays, the ambient electron density is high so that breakdown need not be initiated. As the time delay increases, the ambient electron density decreases, impedence increases, and the voltage required to initiate the current increases. Because they are proportional to the rate of current rise and inversely proportional to the impedence of the tube, the pumping rates decrease with increasing time delay. This behavior is due primarily to the initial conditions set for the pumping pulse by the interpulse afterglow.

Note that at time delays near 20 μs, the rate of current rise is nearly independent of pressures (see Fig. 1). At this short

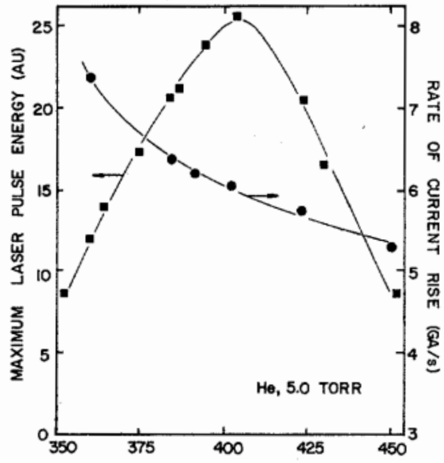

Fig. 4. The rate of current rise at a time delay of 20 μs and laser pulse energy optimized with respect to time delay as a function of tube temperature. The rate of current rise decreases as the CuCl vapor pressure increases.

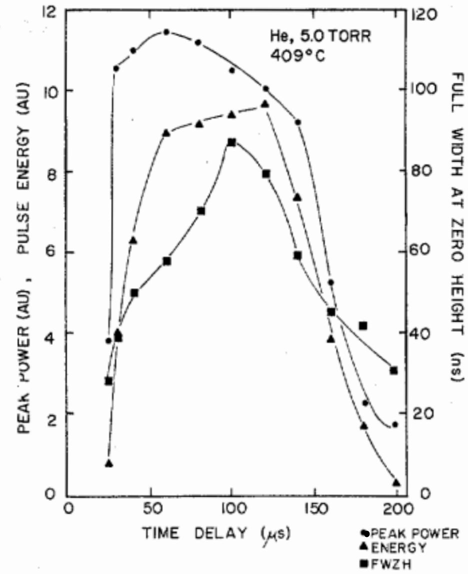

Fig. 5. Laser pulse energy, peak power during the laser pulse, and FWZH as a function of time delay. Note that optimum peak power is obtained at a shorter time delay than optimum energy. Optimum energy is a result of a wider pulse of less than optimum power.

delay, the electron temperature is still relatively high so that little recombination has occurred. The impedence of the tube is therefore small at all pressures. The rate of current rise behaves differently if the buffer gas pressure is held constant and the tube temperature (CuCl vapor pressure) is changed. Fig. 4 shows the rate of current rise at a time delay of 20 μs, and maximum laser pulse energy as a function of tube temperature. The rate of current rise decreases with increasing CuCl vapor pressure. The rate of current rise at the optimum time delay also decreases with increasing tube temperature for temperatures below optimum. Above the optimum tube temperature, the rate of current rise decreases slowly. Pumping rates, therefore, generally decrease as the CuCl vapor pressure increases. Results of a numerical model of the Cu/CuCl laser [9] indicate that the decrease in pumping rates as the tube temperature increases is due in part to a larger rate of electron attachment to free chlorine. This reduces the electron density and reduces the rate of current rise.

The decrease in pumping rates as the tube temperature increases could partly be responsible for the decrease in laser energy observed at high tube temperatures. As the tube temperature increases, the rate at which an inversion can be produced decreases. Hence, despite the potential increase in available copper atoms as the tube temperature increases, laser pulse energy decreases.

The dependence of pumping rates on time delay greatly influences the characteristics of the laser pulse. The shape of the laser pulse, peak laser power, and laser pulse energy all depend on the rate of pumping and time delay. Fig. 5 shows laser pulse energy, peak power, and full width at zero height (FWZH) of the laser pulse as a function of time delay. The behavior is typical for a wide range of buffer gas pressures and tube temperatures. Note that maximum peak power occurs at a shorter time delay than does maximum energy. Maximum energy occurs with a relatively longer pulse having less than the maximum peak power. We have seen that pumping rates decrease as the time delay increases. It appears that maximum peak power occurs at an early delay because the pumping rates are near maximum. Maximum energy, on the other hand, is a result of an optimum density of ground state copper as compared to the density of metastable copper. This conclusion will be discussed in the next section.

III. Analysis Based on Rate Equations

An analysis of the copper laser based on rate equations has been performed. Rate equations for the three copper laser levels and the optical pulse intensity were integrated over a parabolic current pulse. The electron temperature was taken to be $T_e(t) = T_e(0)e^{-t/t_m}$, where t_m is the full width of the current pulse and $T_e(0) = 10$ eV. The details of the analysis are discussed elsewhere [9]. In order to isolate the effect of the rate of current rise on the shape of the laser pulse, the same initial conditions were used for several values of the rate of current rise (2-16 GA/s). Laser output intensity as a function of the rate of current rise is shown in Fig. 6. Note that as this rate decreases, the peak pulse power decreases and the pulse width increases. Remembering that the rate of current rise decreases as the time delay increases, we note that this behavior is similar to that seen experimentally. At short time delays when the rate of current rise is high, peak power is obtained. As the time delay increases and the rate of current rise decreases, the pulse widens. Hence, the experimentally observed shape of the laser pulse is partly explained by the change in the rate of current rise.

Despite the decrease in pumping rates at early delays, the peak power and pulsewidth increases. We believe this initial increase in peak power and pulsewidth are a result of the relaxation of metastable copper which inhibits the laser pulse. This conclusion is based on the results of spectroscopic studies [10] and the results of a detailed model of the dissociation pulse, afterglow, and laser pulse [9]. Although the density of ground-state copper may increase after the dissociation pulse is over [2], our model predicts that laser oscillation will not occur as long as only a few percent of the copper atoms are in

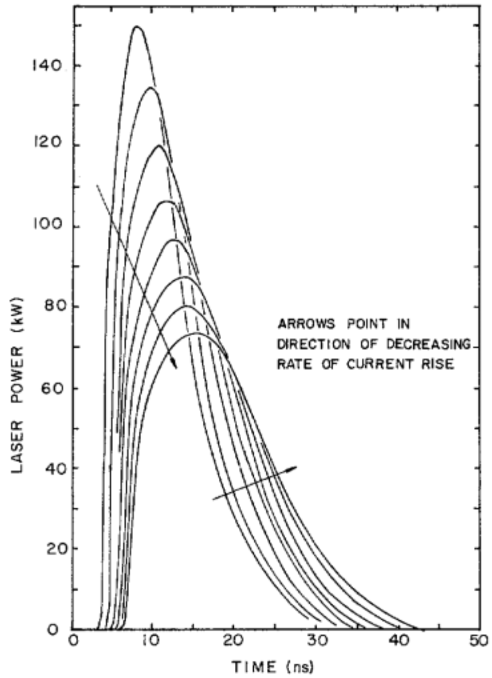

Fig. 6. Typical laser pulses computed using rate equations as a function of the rate of current rise of the pumping pulse. The arrows point in the direction of decreasing rate of current rise (16–2 GA/s). Note that as the rate of current rise decreases, peak power decreases, and the pulsewidth increases.

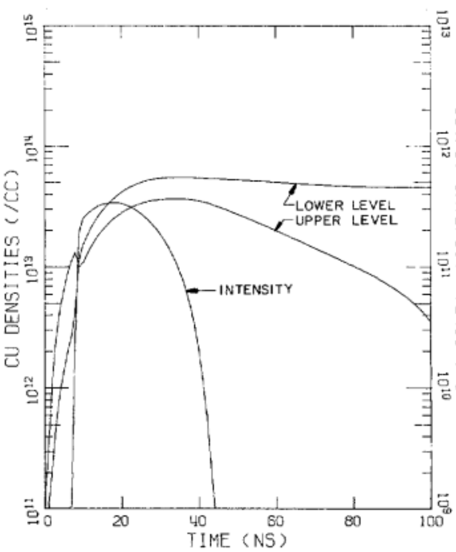

Fig. 7. Laser output intensity, and copper densities for a typical laser pulse as computed with rate equations. The initial electron temperature was 10.0 eV. The peak electron density was $1 \times 10^{14}/\text{cm}^3$.

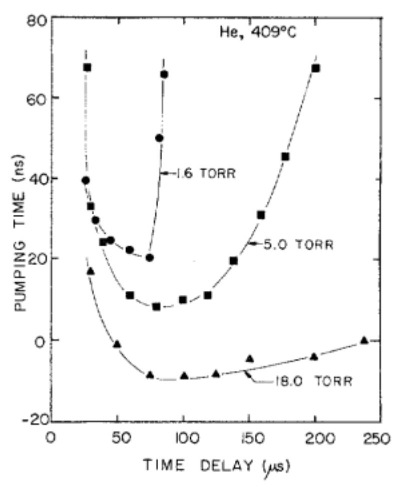

Fig. 8. Pumping time as a function of time delay and buffer gas pressure. The optimum time delays for energy are 40 μs (1.6 torr), 120 μs (5 torr), and 80 μs (18 torr). The optimum time delays for peak power are 35 μs (1.6 torr), 60 μs (5 torr), and 60 μs (18 torr).

the metastable state at the time of the pumping pulse. As the fraction of metastable copper atoms falls below this value, the laser pulse power increases rapidly as a function of time delay. According to these results, the minimum delay and the initial increase in laser power are not a result of an increase in the ground-state density but a decrease in the metastable density.

If the rapid rise in current seen at short delays could be maintained until longer delays, in order to coincide with the optimum densities of ground-state and metastable copper, then maximum peak power and laser energy would be improved. The results of an attempt to do so are reported elsewhere [11].

The dependence of laser energy and power on the rate of current rise can be better understood by referring to Fig. 7. Here we see calculated values of laser intensity and copper densities for a typical laser pulse. The gain reaches a maximum value at which time the cavity intensity attains a sufficiently large value (the critical value) so that stimulated emission rapidly discharges the upper laser level to the lower level. The number density of the upper level drops below that of the lower level but the gain remains positive for a short time due to the favorable degeneracy ratio. Because of this gain switched behavior, laser energy and peak power for a given pulse will be determined largely by the magnitude of the population inversion at the time that the upper laser level discharges to the lower laser level. Once the upper laser level is populated, a finite time is required (due to a finite spontaneous emission coefficient) for the cavity intensity to build up to its critical value. A current which rises faster can more rapidly populate the upper level during this small but finite time. Laser pulse energy and power are then larger.

IV. The Pumping Time Required to Achieve Threshold

With the conclusions of Sections II and III we can now discuss the observed variation in the pumping time required to achieve threshold, as a function of time delay. The pumping time is the time between the initial rise of the pumping pulse current as defined in Fig. 3 and the appearance of the laser pulse. It is a function of time delay, buffer gas pressure, and tube temperature. Fig. 8 shows the pumping time as a function of time delay and buffer gas pressure with the tube temperature fixed. At short delays the pumping time is as large as tens of nanoseconds. As the time delay increases, the pumping time decreases and the laser pulse appears earlier. A minimum pumping time is reached when the laser pulse appears at or near the leading edge of the current pulse. For longer time

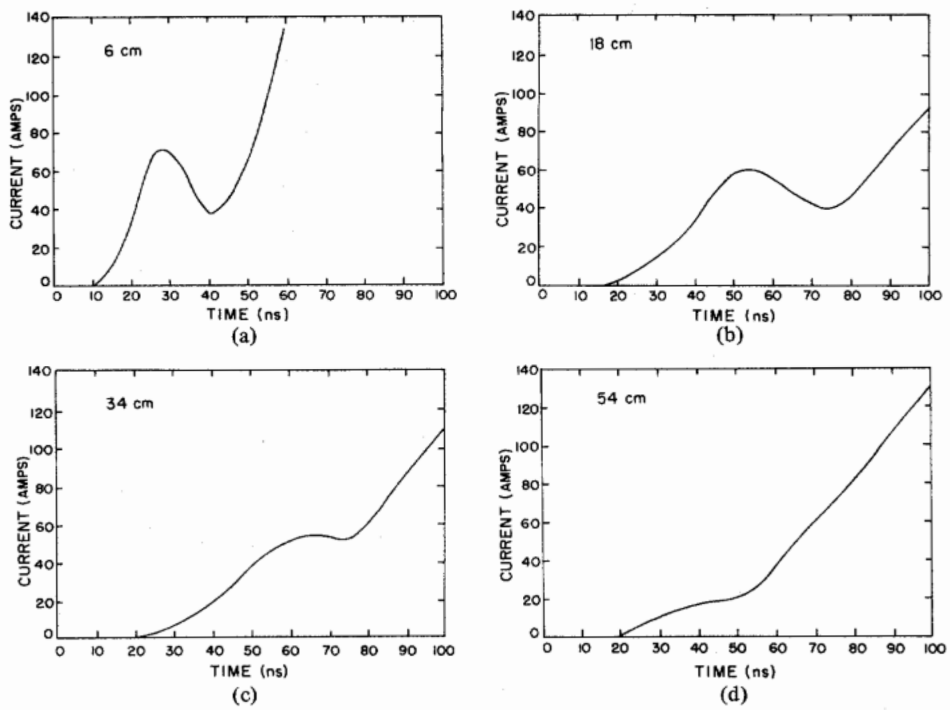

Fig. 9. The leading edge of the second current pulse at a time delay of 40 μs in a double pulse helium discharge. The measurements were made at (a) 6 cm, (b) 18 cm, (c) 34 cm, and (d) 54 cm from the cathode for a discharge tube 60 cm long. The helium pressure was 10.8 torr and charging voltage was 16.0 kV. The main current pulse had a peak current of about 400 A, and a FWZH of about 500 ns.

delays, the pumping time increases and the laser pulse appears later in the current pulse. The delay at which the pumping time is a minimum increases with increasing buffer gas pressure. The minimum pumping time decreases as the buffer gas pressure is increased. The same behavior is observed as a function of tube temperature: for a given time delay, the pumping time decreases with increasing density of CuCl.

The pumping time changes as a function of time delay for the following reasons. At short time delays, there is still a significant population of metastable copper. Hence, a long pumping time is necessary to produce positive gain. As the time delay increases, the metastable population decreases and a shorter pumping time is required despite the decrease in pumping rates. At long delays, despite the lack of metastables, the pumping rates and copper densities are sufficiently small so that a longer pumping time is again required to reach threshold.

Despite lower pumping rates, the pumping time is less at a higher buffer gas pressure. If diffusion and deactivation at the walls were the major loss mechanism for copper atoms in the metastable state, the pumping time would increase with buffer gas pressure. Because the opposite behavior is observed, a volumetric collisional deactivation mechanism must dominate. The similar behavior observed for pumping time when the CuCl density is increased again indicates that collisional deexcitation is the dominant relaxation mechanism.

Note that the minimum pumping time is actually negative for given discharge conditions. (The current transformer and detector have comparable response times.) This indicates that there are excitation processes occuring in the laser tube sufficient to reach threshold before the current pulse can be detected exterior to the tube. The laser tube used in this study is 65 cm long whereas laser oscillation has been obtained for Cu/CuCl lasers in tubes as short as 3 cm [12]. Hence, if the initial increase in electron density consists of an ionization front which propagates from one electrode to the other with application of the voltage, or there are other axially dependent excitation processes which occur, oscillation may start from excited atoms in a small portion of the tube before the current can be detected at the anode [13].

To investigate the possibility of such processes with our discharge circuit, a helium discharge tube was made with an OD small enough to fit through a current transformer. Measurements of current as a function of axial position and time delay indicate that there are precursor current pulses of significant current which precede the primary current pulse and which propagate from cathode to anode. At short time delays, the current increases approximately uniformly along the tube. As the time delay increases, a precursor current pulse develops, first near the cathode, and later nearer the anode (see Fig. 9). The magnitude of the current pulse decreases from cathode to anode. The results of a numerical model of high repetition rate discharges [14] indicate that these precursor pulses are a result of cool electrons which are accelerated by the electric field. The current appears to increase because of an increase in drift velocity and not because of an increase in the electron density. When the electron energy reaches the first excitation threshold and then the ionization threshold, the electrons lose energy at a faster rate and the current appears to decrease. Shortly thereafter, secondary electrons gain energy and the main current pulse develops, changing little as a function of axial position.

Due to the large electron density present when the voltage is applied at short delays, these precursor pulses are probably not the same as the ionization waves previously described [13] which separate a region of low or no ionization from a region of high ionization. The electron density at the front of an ionization wave is $\lesssim 10^{12}/\text{cm}^3$ [13], which is much less than the electron density present within tens of microseconds after a discharge in a noble gas or a metal vapor laser [9], [15].

V. Concluding Remarks

In summary, the rate of current rise and peak current for the pumping pulse in a Cu/CuCl laser have been found to decrease with increasing time delay, increasing buffer gas pressure, and increasing tube temperature. This implies that excitation rates decrease as functions of the same quantities. The causes for this behavior are related mainly to the initial values of electron density, provided by the interpulse afterglow, for the pumping pulse. This conclusion is supported by the observation that larger voltages are required to initiate the pumping pulse current as the time delay increases.

Maximum peak laser power occurs at shorter time delays than does maximum energy. The reason is that maximum power is due to the higher rate of current rise, and therefore more rapid pumping of the upper level, at short time delays, and to the gain-switched nature of the laser. Maximum energy is a result of a wider pulse of less than maximum peak power occurring when the density of ground state copper is near optimum as compared to the density of metastable copper.

The pumping time required for threshold is a function of time delay, buffer gas pressure, and tube temperature. The pumping time is sometimes negative indicating that the excitation processes early during the current pulse are a function of axial position. The variation in this time indicates that volumetric collisional deactivation of the lower laser level is the dominant relaxation mechanism.

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References

[1] C. J. Chen, N. M. Nerheim, and G. R. Russell, "Double-discharge copper vapor laser with copper chloride as a lasant," *Appl. Phys. Lett.*, vol. 23, pp. 514–515, 1973.

[2] J. Tenenbaum, I. Smilanski, S. Gabay, G. Erez, and L. A. Levin, "Time dependence of copper-atom concentration in ground and metastable states in a pulsed CuCl laser," *J. Appl. Phys.*, vol. 49, pp. 2662–2665, 1978.

[3] I. Liberman, R. V. Babcock, C. S. Liu, T. V. George, and L. A. Weaver, "High-repetition-rate copper iodide laser," *Appl. Phys. Lett.*, vol. 25, pp. 334–335, 1974.

[4] W. T. Walter, N. Solimene, M. Piltch, and G. Gould, "Efficient pulsed gas discharge lasers," *IEEE J. Quantum Electron.*, vol. QE-2, pp. 474–479, 1966.

[5] R. S. Anderson, L. W. Springer, B. G. Bricks, and T. W. Karras, "A discharge heated copper vapor laser," *IEEE J. Quantum Electron.*, vol. QE-11, pp. 172–174, 1975.

[6] A. A. Vetter, "Quantitative effect of initial current rise on pumping the double-pulsed copper chloride laser," *IEEE J. Quantum Electron.*, vol. QE-13, pp. 889–891, 1977.

[7] L. J. Kieffer, "A compilation of electron collision cross section data for modeling gas discharge lasers," JILA, Univ. Colorado, Boulder, CO, 1973, Rep. COM-74-11661.

[8] M. A. Biondi, "Recombination" in *Principles of Laser Plasmas*, George Bekifi, Ed. New York: Wiley, 1976, pp. 125–158.

[9] M. J. Kushner and F. E. C. Culick, "A model for the dissociation pulse and afterglow in the Cu/CuCl double pulse laser," *J. Appl. Phys.*, to be published.

[10] N. M. Nerheim, "Measurements of copper ground state and metastable level populations in a copper-chloride laser," *J. Appl. Phys.*, vol. 48, pp. 3244–3250, 1977.

[11] M. J. Kushner and F. E. C. Culick, "A continuous discharge improves the performance of the Cu/CuCl double pulse laser," *IEEE J. Quantum Electron.*, vol. QE-15, pp. 835–837, Sept. 1979.

[12] N. M. Nerheim, "A parametric study of the copper chloride laser," *J. Appl. Phys.*, vol. 48, pp. 1186–1190, 1977.

[13] Ionization waves during breakdown have been observed by a number of investigators. See, for example, A. Haberstich, "Experimental and theoretical study of an ionizing potential wave in a discharge tube," Ph.D. dissertation, Univ. Maryland, College Park, 1965; W. P. Winn "Ionizing space-charge waves in gases," *J. Appl. Phys.*, vol. 38, pp. 783–790, 1966. Ionizing waves have also been treated theoretically. See, for example, G. A. Shelton, Jr. and R. G. Fowler "Nature of electron-fluid-dynamical waves," *Phys. Fluids*, vol. 11, pp. 740–746, 1968.

[14] M. J. Kushner, unpublished, 1979.

[15] V. M. Batenin, V. A. Burmakin, A. I. Evtyunin, I. I. Klimovskii, M. A. Lesnoi, and L. A. Selezneva "Time dependence of the electron density in a copper vapor laser," *Sov. J. Quantum Electron.*, vol. 7, pp. 891–893, 1977.

M. J. Kushner (M'79), photograph and biography not available at the time of publication.

F. E. C. Culick, photograph and biography not available at the time of publication.