Extrema of electron density and output pulse energy in a CuCl/Ne discharge and a Cu/CuCl double-pulsed laser

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Electron-density measurements have been made in a CuCl/Ne discharge using a CO_2 laser interferometer. A local maximum followed by a local minimum in electron density have been found to move a higher tube temperature with higher buffer pressure. Extrema in Cu/CuCl double-pulsed laser energy may be correlated with the extrema in electron density. An analysis based on rate equations yields qualitative agreement with measured laser-pulse energies. The results support the idea that ionization of copper is primarily responsible for the decrease of laser output as the tube temperature is increased.

The copper laser has been the focus of much interest in recent years as a source of intense nanosecond pulses of optical ($\lambda = 5106$ and 5782 Å) radiation.[1-5] Unlike the conventional copper laser which must operate at a tube temperature of about $1500\,°C$ [4] in order to obtain sufficient copper partial pressure from pure copper, the use of copper chloride (CuCl) as a source enables tube temperatures to be within a few degrees of $400\,°C$. The operation of a Cu/CuCl laser requires at least two discharge pulses separated by tens to hundreds of microseconds. The first pulse dissociates the CuCl into Cu and Cl. The second pulse serves as the pumping pulse. Typical optical-pulse duration is 15—30 ns FWHM with a typical output energy of a few millijoules.

The optical-pulse energy of the double-pulsed

Cu/CuCl laser is a sensitive function of tube temperature, geometry, and discharge characteristics.[6] In particular, the pulse energy exhibits a maximum as a function of tube temperature. Measurements of electron temperature for a fixed tube geometry and buffer-gas pressure[7] revealed that the average electron energy is a minimum at tube temperatures for which the optical-pulse energy is a maximum. In a continuing effort to understand the operation of the Cu/CuCl laser, this letter reports results of electron-density measurements in a CuCl discharge taken with a CO_2 laser interferometer.

The CuCl discharge was sustained in a 2.54-cm i.d. quartz tube enclosed in a 46-cm-long heater; the electrode spacing was 35.5 cm. The electric discharge con-

sisted of a breakdown pulse (14.4-nF capacitor charged
to 5.0 kV) immediately followed by the discharge of a
60-μF capacitor charged to 1.5 kV. Copper chloride
was placed in a trough running the length of the tube,
and the tube was filled with neon. The neon was cir-
culated past NaCl windows to avoid deposition of CuCl
on the colder window surfaces. The CO_2 laser inter-
ferometer, operating in the TEM_{00} mode at 10.6 μm,
was mounted on a granite table to minimize vibration.
The interferometer was constructed according to the
descriptions given in Refs. 8 and 9. The electron den-
sities measured with the interferometer were taken as
the maximum electron densities occurring during the
discharge pulse.

The results of the electron-density measurements
are shown in Fig. 1 for four Ne buffer-gas pressures.
For a fixed pressure of buffer gas, there appears both
a local minimum and maximum in electron density as
the tube temperature is changed.[10] The tube tempera-
tures at which these extrema appear increase with in-
creasing buffer-gas pressure. The pattern consists of
a maximum followed by a minimum at a higher tube
temperature. When Ne alone was placed in the dis-
charge, the local minimum in electron density did not
appear. The maximum, however, was found at the
same tube temperature for a given pressure as in the
CuCl/Ne discharge.

The increase in electron density at low tube temper-
atures can in part be explained by an increasing E/N
and neon ionization coefficient[11] as the tube tempera-
ture rises $(N \sim 1/T)$. At higher temperatures diffusion
losses of electrons to the walls become dominant and
the electron density is reduced. As the CuCl vapor
pressure becomes about 1% of the total pressure, the
Cu and Cl atoms with their lower ionization potentials
contribute heavily to the electron population, thereby
increasing the electron density.

The same discharge tube was used as a Cu/CuCl
laser. The excitation load was changed to a double-pulse
arrangement[12] operated at 12.5 kV and employing

FIG. 2. Experimental and computed results for laser-pulse
energy at a tube temperature of 390/C. The experimental
scale was chosen for ease of comparison.

14.4- nF dissociation and 14.4-nF pumping capacitors.
The laser-pulse energy was optimized with respect
to the time delay between dissociation and pumping
pulses. A photodiode was used to measure the optical-
pulse energy. For experimental convenience, the tube
temperature was held constant and the Ne pressure
was varied. The results for a tube temperature of
390 °C are shown in Fig. 2. The laser-pulse energy
reaches a maximum at low gas pressure, shows a local
minimum as the pressure is increased, and then de-
clines to low values at higher buffer-gas pressure. As
shown in Fig. 3, a correlation can be made between
the extrema in electron density and the extrema in
laser-pulse energy. When plotted as a function of tube
temperature, the pressures at which the electron den-
sity is locally minimum and laser-pulse energy is
maximum fall approximately on a straight line. Similar
behavior was found for the pressures at which the elec-
tron density is a local maximum and laser-pulse energy
a local minimum. This correlation between maximum
laser output energy and minimum electron density has
also been made with the positive-column He-Cd⁺ laser.[13]

These results, when combined with the results of
the electron temperature measurements,[7] suggest the
following interpretation. At higher electron densities
$(n_e \approx 2 \times 10^{14}/cm^3)$ and correspondingly high electron
temperatures $(T_e \approx 15$ eV), the copper ionization rates
from both ground state and excited levels exceed the
excitation rates to the upper laser levels. The laser-
pulse energy is therefore depressed because there is
less ground-state copper available to pump to the upper
level. Charge exchange with neon ions and Penning
ionization with neon metastables during the afterglow
between dissociation and pumping pulses will also con-
tribute to the depletion of ground-state copper. Due to
the high ionization potential (21.6 eV) and metastable
energy (16.7 eV) of neon, these affects will be impor-
tant only at the higher electron temperatures referred
to above. As the electron density and electron temper-
ature decrease, the ionization rates are reduced,
leaving more ground-state copper, and the excitation
rates are increased. At these electron temperatures
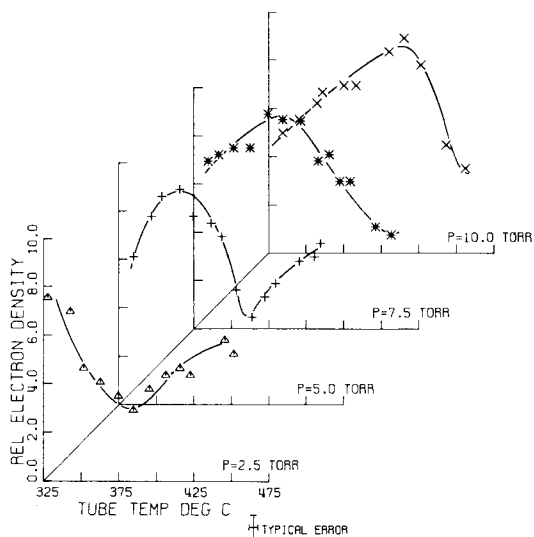$(T_e \sim 10$ eV), the electron distribution is maximum near

FIG. 1. Electron densities in a pulsed CuCl/Ne discharge.
With increasing pressure, both the minimum and maximum
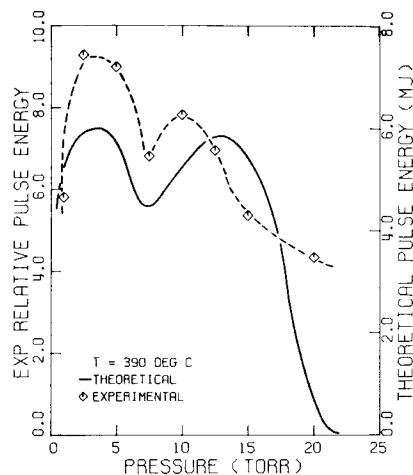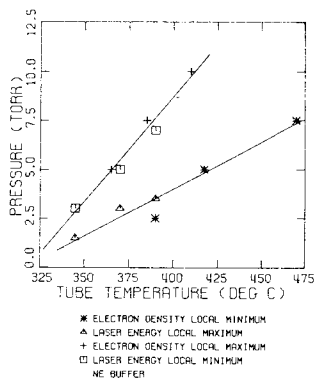in the electron density shift to higher temperature.

FIG. 3. When plotted as a function of tube temperature, pressures at which electron density is a minimum and laser energy is a maximum fall on a straight line. The same is true for electron density maximum and laser energy minimum.

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※ ELECTRON DENSITY LOCAL MINIMUM
△ LASER ENERGY LOCAL MAXIMUM
＋ ELECTRON DENSITY LOCAL MAXIMUM
□ LASER ENERGY LOCAL MINIMUM
  NE BUFFER
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the peak of the resonance-impact cross section for excitation to the upper laser levels.[14,15] Under these conditions, laser-pulse energy is a maximum. At low electron densities ($n_e \lesssim 5 \times 10^{13}$) and temperatures ($T_e \approx 5$ eV), the ionization rate is reduced significantly so that there is an abundance of copper atoms available for excitation, but because the optimum electron temperature for excitation to the upper laser level occurs at higher values while the excitation rate to the lower laser level is becoming larger, the net excitation rate is lowered and the laser pulse energy is also small.

With the above interpretation as a basis, a set of rate equations have been constructed for the copper laser. Five equations are used for the populations of three copper atom levels and two neon levels; the sixth equation governs the optical-pulse intensity. As a simplification, the populations of CuCl and Cl are not considered. Semiempirical cross sections for the excitation of copper[14,15] and an experimental electron distribution function[16] are used. The value of the electron density at the peak of the discharge current pulse was specified for each tube temperature and buffer-gas pressure in accordance with the experimental results reported here. The assumption is made that the electron temperature is constant for a given tube temperature and pressure and directly proportional to the peak electron density. The rate equations have been integrated over a 100-ns-wide parabolic current pulse using the Runge-Kutta-Gill method with Adams-Moulten correction features.[17] Typical results for one set of experimental conditions are shown in Fig. 2.

These results reproduce the observed extrema in laser energy. Gryzinski cross sections[18] were initially used for copper ionization rates, but it was found that the peak value of these cross sections had to be increased by at least a factor of 3 in order to obtain the results displayed. The behavior observed at high pressures ($p > 10$ Torr) is not represented well. In particular, the second maximum in laser energy is displaced to higher pressures and the value of the second maximum is too high, being nearly equal to that of the first. This discrepancy may be partly due to the difference at higher pressures between the CuCl discharge, which provide the values for the electron density, and the double-pulsed discharge used for the laser.

Because the differences are greater at high pressure, one would expect that processes involving the buffer gas

may be responsible for the reduced optical-pulse energy. With this in mind, Penning ionization rates ($Ne^* + Cu \to Ne + Cu^+ + e$) and the initial fraction of metastable neon were varied in the model. A Penning cross section of 5×10^{-15} cm^2 and initial metastable fractions of 0.1 were required in order to reduce the high-pressure optical-pulse energy to experimental values. The relative influence of the slower atom-atom interaction rates on the optical-pulse energy is small because the laser pulse occurs in the first 40 ns of the current pulse. The laser pulse is over before the population of excited neon has reached maximum. It appears that one must look to atom-atom processes occurring in the relatively long afterglow between the dissociation pulse and the pumping pulse to explain the high-pressure behavior.

In summary, electron densities have been measured in a CuCl/Ne discharge using a CO$_2$ laser interferometer. Local maximum and minimum values of electron density were found to move to higher tube temperature at higher buffer pressures. The maximum values of electron density were found to correlate with minimum values of pulse energy from a double-pulse Cu/CuCl laser. Minimum values of electron density were found to correlate with maximum values of pulse energy. An analysis based on rate equations using experimental values of electron density and temperature as input data reproduced the extrema in the pulse energy. This agreement between calculated and experimental results supports the proposition that ionization of copper is a dominant mechanism in reducing the pulse energy for a double-pulsed Cu/CuCl laser operated at tube temperatures greater than the optimum value.

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