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Discharge studies of the Ne-Cu laser[a]

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Spontaneous-emission and absorption studies of the Ne-Cu hollow-cathode laser are summarized. The major discharge processes operative in the Ne-Cu laser are outlined and the qualitative aspects of a proposed model of the Ne-Cu laser are discussed. Emphasis is placed on cathode sputtering as a source of copper atoms, and we demonstrate that copper densities of 10^{14} atoms/cm^3 are created via discharge sputtering.

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We have reported laser action, excited in hollow-cathode discharges, originating in the ion systems Cu II, Ag II, and Au II at wavelengths ranging from 220.0 to 840.0 nm.[1–3] It is believed that the upper laser levels are populated via charge transfer in a reaction of the form

$$R^+ + M \rightarrow R + M^{+*} + \Delta E \qquad (1)$$

where R^+ is the ground-state He or Ne ion, M^{+*} is an excited metal ion, and ΔE (kinetic energy) is the energy difference between R^+ and M^{+*}.

A practical advantage of the hollow-cathode-laser design is that the metal vapor is generated via cathode sputtering rather than by an external oven or by discharge heating. To evaluate the sputtering mechanism quantitatively, we have measured the vapor density of sputtered copper as a function of discharge current and neon pressure in a Ne-Cu hollow-cathode discharge. Moreover, by studying the spontaneous emission from selected levels in the neutral and singly ionized spectra of both neon and copper, we now understand the major discharge processes occurring in the Ne-Cu hollow-cathode laser.

The hollow cathodes used for spectroscopic measurements were 20 mm in length and 2×6 mm in cross section and identical to those employed in actual laser tubes, but of shorter length (see Refs. 1—3). The tubes were water cooled and could sustain a transverse dc current of 0.5 A/cm. The threshold current for laser action is typically 0.1 A/cm. Since absorption as well as emission studies were made, two *identical* tubes were placed in tandem; one was a source of line radiation for absorption studies and the other was the absorber. The ground-state copper density and the neon metastable densities, were determined using the analysis pioneered by Ladenburg and Reiche.[4] Note that the hyperfine splitting of the copper ground state[5] exceeds the Doppler width by a factor of 8 and was included in the interpretation of our absorption measurements.

Copper atom densities measured in the center of the 2×6-mm cathode slot as a function of discharge current are displayed in Fig. 1. For currents between 0.01 and 0.05 A/cm, the copper density increases nonlinearly with current, showing an I^n dependence with $n \approx 3.8$. This nonlinear dependence has also been observed by other investigators in experiments on the sputtering yield of metals in low-current glow discharges.[6,7] However, above 0.05 A/cm, our measurements show

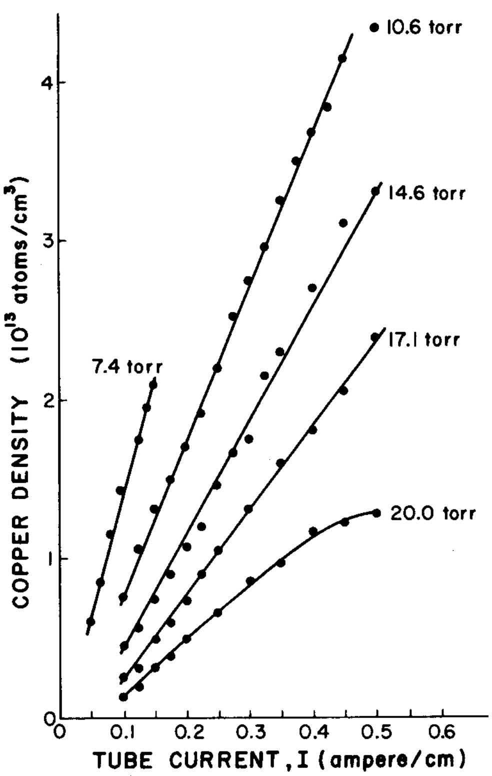

FIG. 1. Sputtered copper-vapor density as a function of discharge current and pressure measured in the center of the 2 ×6-mm cathode slot.

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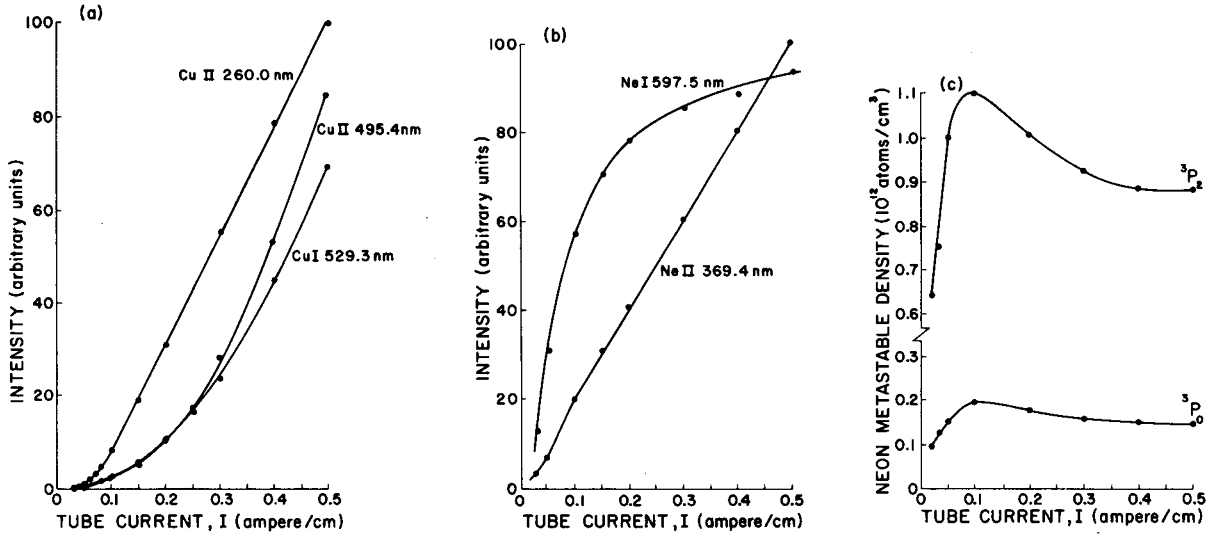

FIG. 2. (a) Current dependence of the Cu I and Cu II spontaneous emission at 10.6 Torr (3.2×10^{17} atoms/cm^3) neon fill. (b) Current dependence of the Ne I and Ne II spontaneous emission at 10.6 Torr (3.2×10^{17} atoms/cm^2) neon fill. (c) Neon metastable densities versus discharge current at 10.6 Torr (3.2×10^{17} atoms/cm^3) neon fill.

that the copper density varies in a *linear* manner with increasing discharge current. At a neon pressure (density) of 10 Torr (3×10^{17} atoms/cm^3) and at a discharge current of 0.5 A/cm, the copper density reaches 4×10^{13} atoms/cm^3. Spatially resolved absorption measurements (resolution 0.15 mm) show that the copper density near the walls of the slot is about twice as high as that measured in the center. Thus, in our Ne-Cu hollow-cathode discharge, densities up to 10^{14} atoms/cm^3 are realized. To obtain this copper density via thermal generation, a temperature in excess of 1200 °C is required.[8] The effect of increasing neon density on the sputtered copper density is also shown in Fig. 1. Note that the copper density increases as the neon pressure decreases in qualitative agreement with the sputtering theory of Guntherschulze.[9]

The current dependence of the Cu I and Cu II spontaneous emission is shown in Fig. 2(a). The intensity of spontaneous emission of Cu I lines varies quadratically with discharge current, as is illustrated by the 529.3-nm Cu I transition. The behavior of Cu II spontaneous emission falls into two categories. Spontaneous emission from all Cu II *laser* transitions is strong and is observed to vary *linearly* with increasing discharge current. Since the major source of ground-state copper ions arises from radiative decay of charge transfer excited Cu II levels, we believe that the ground-state Cu II density also varies linearly with discharge current. Note also in Fig. 2(a) that the spontaneous emission of copper transitions, which is most distinct for Cu II laser transitions, displays a distinct threshold behavior at 0.05 A/cm. This current threshold increases with increasing neon pressure. On the other hand, the spontaneous-emission intensity from Cu II levels which do *not* support laser action is weak and varies nonlinearly with discharge current. Consider the 495.4-nm transition of Cu II whose upper level lies more than 26 000 cm^{-1} above the energy of Ne$^+$ and, hence, cannot be excited by reaction (1) in a Ne-Cu discharge. The intensity of this transition varies roughly quadratically with increasing discharge current, as shown in Fig. 2(a).

The measured Ne I and Ne II spontaneous-emission intensity versus discharge current is summarized in Fig. 2(b), while Fig. 2(c) displays the variations of the neon ^3P metastable densities with discharge current. The Ne I spontaneous-emission intensity, as illustrated by the behavior of the 597.5-nm transitions, shows a tendency towards saturation at high current density. In sharp contrast, Ne II transitions vary *linearly* (above 0.10 A/cm) with increasing current [see Fig. 2(b)]. Finally, from Fig. 2(c), it can be seen that the neon metastable density does *not* vary with current above 0.15 A/cm. The above observations are consistent with a Ne-Cu discharge model presented below.

Under our experimental conditions and geometries, the Ne-Cu hollow-cathode discharge is practically identical to that described by White.[10] A negative-glow plasma develops in the slot of the cathode, and this glow is separated from the cathode surface by a sheath which is much smaller than the cathode dark space in a normal glow discharge. The greater part of the discharge voltage V_c develops across the sheath, whereas the negative-glow region is field free. We observed, as did White,[10] that the tube voltage (270–300 V) does not vary appreciably with discharge current over the range 0.025–1.0 A/cm. To a first approximation then, the energy with which the electrons enter the negative glow is eV_c and is *not* strongly dependent on the discharge current. Hence, as the discharge current is increased, the characteristic electron temperature of the discharge is *unchanged*, and the electron density is expected to vary linearly with current.[11]

A mathematical model of the Ne-Cu discharge is presented in quantitative detail elsewhere.[12] Only the qualitative aspects of this model are outlined here to explain our experimental observations. First, the observed quadratic behavior with current of the Cu I and Cu II (nonlaser only) spontaneous-emission intensity may be explained from a linear increase of both the electron density (see above) and the copper-vapor density (measured). Hence, if the dominant excitation mechanism of these lines is electron impact, a quadratic behavior results. We cannot determine whether the dominant excitation of the nonlaser Cu II lines is via electron impact on ground-state neutral atoms or on ground-state copper ions. Both mechanisms give the observed quadratic behavior. For all Ne I lines observed, the less than linear increase of the spontaneous-emission intensity is associated with the competition between cumulative ionization and radiative decay. The behavior of the 597.5-nm Ne I transition is illustrative and is shown in Fig. 2(b). The observed linearity of the Ne II spontaneous emission [Fig. 2(b)] is consistent with single-step ionization and excitation of neon atoms by electrons. Note, however, that since the neon-ion density is likely to be independent of discharge current (see below), one cannot distinguish between single- and multiple-step electron impact excitation from the Ne II spontaneous-emission behavior.

The spontaneous emission from Cu II levels which support laser action is explained as follows. In our model,[12] the rate equation for neon ions is

$$\frac{d}{dt}[\mathrm{Ne}^+] = k_1[n_e][\mathrm{Ne}] + k_2[n_2][\mathrm{Ne}^*] - k_3[\mathrm{Ne}^+][\mathrm{Cu}]. \qquad (2)$$

[Ne], [Ne*], and [Ne⁺] represent the densities of the neon ground state, various excited neon states (predominantly metastables), and the neon-ion ground state, respectively; $[n_e]$ and [Cu] represent the electron density and ground-state copper density; and k_1, k_2, and k_3 are rate constants. The first term on the right side of Eq. (2) represents direct electron impact ionization of neon atoms. This term varies linearly with discharge current since the electron temperature is assumed constant with increasing current. The second term represents the production rate of neon ions via cumulative ionization. To a first approximation, the greatest contribution to cumulative ionization is from electron collisions with neon metastables. Because we have measured the densities of the neon $^3P_2$ and $^3P_0$ metastables to be *constant* with increasing discharge current ($I > 0.1$ A/cm), cumulative ionization will also vary linearly with current. The major loss process for neon ions, term three, is assumed to be via reaction (1). Since the measured copper density varies linearly with current, all terms on the right-hand side of Eq. (2) contain $[n_e]$. Hence, the steady-state neon-ion density is *independent* of discharge current and, according to reaction (1), a *linear* behavior of the spontaneous emission from Cu II laser levels is predicted.

The observed threshold for spontaneous emission of Cu II laser transitions, as shown in Fig. 2(a), is also predicted by our model of the Ne-Cu discharge.[12] The model includes the sputtering contributions from *both* copper and neon ions. For incident ion energies below 500 eV, the sputtering yield of a cathode surface varies drastically with incident ion energy and to a lesser extent with ion mass.[13] We believe that the copper ions hit the cathode surface with nearly the full cathode fall (eV_c), whereas the neon ions strike the cathode surface with only a fraction of the full cathode fall energy. This large difference in ion energies occurs because the estimated mean free path[14] for copper ions in neon is 1 mm, $Q(\mathrm{Ne\text{-}Cu^+}) \cong 10^{-16}$ cm^2. However, the mean free path[14] for neon ions in neon is 0.1 mm, $Q(\mathrm{Ne\text{-}Ne^+}) \cong 10^{-15}$ cm^2. Note that copper ions do not experience a significant number of resonant charge transfer oscillations because the copper density is 10^4 times less than the neon density. To a first approximation then, resonant charge transfer collisions determine the energy of the neon ions via the usual E/p relationship, whereas the copper ions experience a nearly free-fall behavior. As a consequence, energetic copper ions play a crucial role in our sputtering model.

Using the sputtering data accumulated by Wehner[13] and our estimated energies for copper and neon ions impinging upon the cathode surface, the ratio of the sputtering yield of a copper ion (γ_c) to that of a neon ion (γ_N) is approximately 100. Based on this two-order-of-magnitude difference in sputtering yield, our quantitative discharge model predicts that the current dependence of the copper density within the hollow cathode will show three distinct regions. At low discharge current the sputtering is dominated by inert-gas ions striking the cathode surface with a yield proportional to γ_N, whereas at high current the sputtering is dominated by metal ions with a yield γ_c. At intermediate currents, the dominant species in the cathode ion current changes from neon ions to copper ions. This transition of ion species from neon ions to copper ions is due to reaction (1) and causes the observed threshold behavior of Fig. 2(a). According to our quantitative model, the sharpness of the transition region is a function of the ratio of sputtering yields γ_c/γ_N. Our model also predicts that at high current a linear relationship exists between the copper density and the discharge current, which is consistent with the density measurements summarized in Fig. 1. Note that in Fig. 1, the change in slope of the copper density plots is interpreted, according to our model, as the variation of γ_c with ion energy, and γ_c reaches a maximum at lowest neon pressure, in agreement with the empirical sputtering laws first proposed by Guntherschulze.[9] Complete details are given elsewhere.[12]

In summary, we have measured the density of sputtered copper atoms in the Ne-Cu hollow-cathode laser discharge as a function of discharge current and neon pressure. The major discharge processes in the Ne-Cu hollow-cathode laser are outlined. Measurements of spontaneous emission from the various species present in the discharge are consistent with a proposed discharge model which includes sputtering.

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