High-Gain Hollow-Cathode Metal Ion Lasers for the UV and VUV

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Abstract—The segmented hollow-cathode pumping discharge gives a higher gain on the He–Cu$^+$ 781, He–Au$^+$ 282, and Ne–Cu$^+$ 270-nm lines at lower discharge current and input power than do the conventional hollow-cathode, hollow anode cathode and helical hollow-cathode discharges. The segmented hollow-cathode offers the prospect of compact low-cost repetitively pulsed UV laser for applications to biomedical and other spectroscopically-based instrumentation. The projections for increasing the gain on the He–Cu$^+$ VUV lines indicate that laser action on one or more of these lines may be achieved.

I. INTRODUCTION

The known charge-transfer pumped transitions in He–Au$^+$, He–Ag$^+$, and Ne–Cu$^+$ provide the basis for a compact low-cost laser emitting a continuous-wave, UV (≈220–290 nm) TEM$_{00}$ beam at the milliwatt level. Such a laser would find many applications in biotechnology and other spectroscopically-based instrumentation. For example, the He–Au$^+$ 282 nm laser would be well suited as a repetitively pulsed light source in an instrument for monitoring biopolymers in electrophoretic capillary channels with at least a sub-nanomole detection sensitivity with the potential to approach the femtomole level. For this and other envisioned applications, repetitively pulsed operation at a low duty cycle and at low peak power is adequate. Existing devices, such as the Optical Parametric Oscillator and the Argon Ion Laser with Intracavity Frequency Doubling, are expensive and are therefore unsuitable as economic light sources for the above instruments. Semiconductor diode pumped solid state lasers, such as the Nd:YAG with frequency quadrupling to give a 265-nm beam, represent the most significant competitive technology in the longer term. Moreover, in relation to intracavity frequency doubling techniques for VUV generation, the best frequency doubling materials (BBO and LBO) are unsuitable since they have significant optical absorption at wavelengths below 190 nm.

The first purpose of this paper is to review the existing studies of the above laser transitions, to identify the important issues that bear on the development of a practical laser, and to present a progress report on the results obtained on the He–Cu$^+$ 781, He–Au$^+$ 282, and Ne–Cu$^+$ 270-nm laser lines using a novel high-voltage hollow-cathode pumping discharge. A second purpose is to discuss the potential for using a scaled-up version of the laser tube to achieve CW laser emission on the He–Cu$^+$ VUV 6$s$ → 4$p$ (~150–160 nm) transitions for the first time. The upper laser level for the Cu$^+$ 781 nm transition is common to a number of strongly pumped Cu$^+$ transitions in the VUV, Fig. 1. The He–Cu$^+$ infrared line, which is of high gain, is consequently both important and convenient for optimizing the pumping of the VUV lines. The Ne–Cu$^+$ UV 5$s$ → 4$p$ (249–272 nm) transitions are also shown in Fig. 1.

II. HOLLOW-CATHODE AND HIGH-VOLTAGE VARIANTS

Metal ion laser transitions pumped by thermal charge transfer were first demonstrated using a conventional hollow-cathode (HC) discharge to generate both the metal vapor by sputtering and the rare-gas ions required to pump the specific metal ion transitions. The two most common geometries for a HC discharge are shown in Fig. 2(a). The process of thermal charge transfer is represented by the following equation

$$B^+ + M \rightarrow B + M^{++} + \Delta E,$$

where $B$ and $B^+$ denote a buffer gas atom and ground state ion respectively and $M$ and $M^{++}$ represent a ground state
metal atom and ion in an excited level. The cross section for charge transfer attains a maximum value for a given buffer gas ion and metal atom combination when the excited metal ion level lies at \( \Delta E \sim 0.2 \) eV below that of the ground state buffer gas ion [1]. The pumping rate for the charge transfer process is proportional to the product \([B^+][M]\). The primary advantages of the HC discharge are firstly, that a high degree of ionization is produced in the negative glow region by virtue of the oscillation of the beam electrons in the potential well between opposed faces of the cathode [2], and secondly that a high density of buffer gas ions can be maintained in the rare-gas/metal vapor mixture despite the high concentration of metal vapor [3]. The early HC sputtering laser work, reviewed in [4], was very encouraging in that beam power at wavelengths in the 224 to 290 nm range as high as 500 mW was obtained from these devices using an active length of 100 cm. Subsequently a power of the order of 20 mW was achieved using a shorter active length of 20 cm [5]. Values of the small-signal gain for such lasers are typically in the range 2–11% m⁻¹ [6]. However, these HC devices invariably required a high operating current and exhibited a threshold current of the order of 10 A, the discharge voltage being about 300 V. Typically, the operating current density \( (J) \) is of the order of 500 mA cm⁻².

The operation of a Ne–Cu⁺ slotted HC sputtering discharge was successfully modeled by Warner et al. [7]. They showed that the neon ion concentration \([Ne^+]\) saturates at a relatively low current density \((\sim 10 \text{ mA cm}^{-2})\) due to the balance between the rate of generation of the ions and their loss through the dominant process of thermal charge transfer to the metal atoms. That study also showed that, although the sputtering process at low current densities is due mainly to rare-gas ions, there is a transition to metal ion dominated sputtering as the current density is raised. In each of these regimes the concentration of ground state Cu atoms \([\text{Cu}]\) rises linearly with the current density at the same rate. The model was used to fit a curve for \([\text{Cu}]\) versus current, as measured by atomic absorption spectroscopy. This gave an estimate of the rate coefficient for the Ne⁺–Cu charge transfer, and the sputtering yield for each of the Ne⁺ and Cu⁺. However, no measurements of \([\text{Ne}^+]\) and \([\text{Cu}^+]\) were presented. Koch and Eichler [8] subsequently applied the Warner model to a similar discharge and showed that estimates of \([\text{Ne}^+]\), \([\text{Cu}^+]\), and \([\text{Cu}]\) and the sputtering yields for the Ne⁺ and Cu⁺ can be derived from a measurement of the dependence on the current of the spontaneous emission on the charge-transfer pumped Ne⁺–Cu 260-nm line, although no independent measurements of these quantities were given. Despite there being significant disagreement between the estimates of the sputtering yields given in the above papers, both agree that there is a saturation of \([\text{Ne}^+]\).

The pumping rate for the laser transition is proportional to the product \([\text{Ne}^+]\cdot [\text{Cu}]\) so that, following saturation of \([\text{Ne}^+]\), it scales linearly with the current density. It should be noted that the use of a single discharge to generate both \([\text{Ne}^+]\) and \([\text{Cu}]\) implies that they cannot be varied independently.

The high threshold current for the HC device is a major obstacle to the development of a compact sputtering-based ultraviolet laser pumped by thermal charge-transfer. The underlying technical problems of vapor containment, cathode erosion and deposition of metal vapor on insulators and the laser mirrors and depletion of rare-gas atoms due to gettering by condensing metal vapor, directly limit the tube lifetime. The lifetime issues for a thermally loaded hollow-cathode CW metal ion laser emitting on visible and near infrared lines has been thoroughly addressed [9]. As yet, a few studies have been made of the factors limiting the lifetime of the sputtering-based HC laser [10]–[12], but these have not addressed the issue of sealed-off operation. However, it has been shown that the rate of erosion of a helical copper cathode [see Fig. 2(c)] is proportional to \(J^{5/2}\) [13] and a 1000 hour cathode lifetime...
has been projected for a current density of 200 mA cm\(^{-2}\). It is clear that the problem of achieving a satisfactory tube lifetime will be eased considerably by a significant reduction in the current density required to reach threshold. To this end, it is important to increase the efficiency of the discharge for pumping by the charge-transfer mechanism. The key to raising the efficiency of the discharge is to modify the hollow-cathode to give a higher voltage and slope resistance, whilst retaining the oscillation of the beam electrons. This can be achieved by increasing the rate of loss of ions and electrons at a boundary of the negative glow. A higher operating voltage is required to compensate this loss by increasing the rate of ion production in the negative glow. The higher discharge voltage results in a more favorable electron energy distribution with an increased concentration of fast electrons and enhances the sputtering due to the increased energy of the ions impinging on the cathode surface. The higher slope resistance increases the stability of the discharge against the formation of arcs.

The first demonstration of this principle used a hollow anode cathode (HAC) discharge [14], [15]. A cylindrical array of anode rods was placed concentrically within and close to a cylindrical hollow-cathode to give a partially obstructed discharge. However, despite the innovation of a slotted thin-walled tubular anode [16], [Fig. 2(b)], the anode of the HAC acts as a sink for sputtered metal vapor and restricts the useable volume within the hollow-cathode. In an alternative design, the helical hollow-cathode (HHC) was employed [17] and used to pump the He-Cu\(^+\) 781-nm [18] and the Ne-Cu\(^+\) 248–270-nm [19] lines. In this device, [Fig. 2(c)], the voltage for a given current is raised by increasing the pitch of a helical coil cathode. For a current density similar to that used in a HC laser discharge, the voltages for the HAC and HHC devices are in the range 500–1500 V, depending on the geometry and the buffer gas and its pressure.

The performances of the HAC and HHC lasers were shown to be superior to that obtained using the HC discharge. The efficiency of the discharge is increased because the buffer gas ion concentration \(\text{[B\(^+\)]}\) will saturate at a higher level and because the metal atom concentration \(\text{[M]}\) rises with current due to both the increase in the sputtering ion flux and in the ion energy (hence a higher sputtering yield). It is appropriate to note that the studies so far made on the HC, HAC, and HHC lasers have emphasized achieving a beam power of the order of 300 mW and have required active lengths greater than 50 cm, so that the threshold current is typically in excess of 10 A. The existing HC and its variants have suffered from the disadvantage of poor vapor containment and a complexity in structure which has limited the scope for tube assembly using metal ceramic vacuum seals. The use of the latter is an essential step toward the elimination of organic vapors and the production of a bakeable sealed-off tube with internal mirrors.

The segmented hollow-cathode (SHC) is a high-voltage hollow-cathode [20] which avoids interposing anodes or other surfaces within the cathode. The anodes and cathodes are arranged around a common cylindrical surface which defines the bore of the discharge tube. Although the anode surfaces act as sinks for the metal vapor, the potential exists for commutating the electrodes. In addition this geometry and structure are compatible with the assembly of a discharge tube using metal ceramic seals. Fig. 2(d) shows the cross section of the SHC discharge tube used to obtain the experimental results described in this paper. The beam electrons can oscillate between and are focussed by the opposed cathode segments. The discharge voltage for a given current density is raised compared with that for a conventional HC because the adjacent anode surfaces introduce an increased localized loss of ions and electrons from the negative glow. Fig. 3 shows a comparison of the voltage-current characteristic for the tube using two opposed segments as a SHC (Fig. 2) with that obtained when all four segments are connected to form a HC. The active length and tube bore are 5 cm and 4 mm, respectively, and the buffer gas He-4% Ar at a pressure of 15 mbar which is near optimum for the operation of the He-Cu\(^+\) 781-nm laser transition. The high gain of the laser tube allowed operation using Brewster windows of fused silica and external mirrors. This had the advantage that the small-signal gain could be measured directly by introducing a calibrated loss into the cavity. However, since the tubes were assembled using vacuum grade epoxy to seal the metal and ceramic parts, bakeout by operation at temperatures above 100 C was not practicable. The laser was pulsed at 1 ps (duration ~5 mS) to allow quasi-CW operation. The following sections summarize laser performances for the SHC on the He-Cu\(^+\) 781-nm, He-Au\(^+\) 282-nm, and Ne-Cu\(^+\) 270-nm lines. The bore diameter is 4 mm unless otherwise stated.

### III. He-Cu\(^+\) 781-nm Laser

A comprehensive parametric study of the operation of the SHC tube on the He-Cu\(^+\) 781-nm transition has been made [20]. The SHC laser has also been shown to have a lower threshold and a superior scaling of the output compared with
the HC and the HAC devices when operated on this line [21]. The dependence of the small-signal gain on the current is shown in Fig. 4. The gain is measured by inserting a calibrated loss (two contra-rotating fused silica plates) into the laser cavity. One or more modules are used to balance the cavity losses, including that of the attenuator when the plates are set at the Brewster angle. The module(s) for which the gain is to be measured are then pumped and the insertion loss increased to restore the laser to threshold. A gain of about 85% m\(^{-1}\) is obtained for a linear current density of 480 mA cm\(^{-1}\). For a fixed linear current density, the gain is approximately doubled when the bore diameter is reduced from 4 to 2 mm [22]. This result is highly significant for the following reasons. First, a reduced diameter is conducive to restricting operation to the TEM\(_{00}\) mode. Second, a given current density (mA cm\(^{-2}\)) requires a reduced current. Finally, the higher gain at small diameters allows the use of a shorter active length.

IV. He–Au\(^{+}\) 282-nm LASER

This transition was shown to have a relatively low threshold current when operated using a HC with internal mirrors [5]. To the best knowledge of the authors, the first measurement of the small-signal gain was that made using a SHC and the intracavity calibrated loss technique [23]. The dependence of the gain on the discharge current (Fig. 4) shows that a gain in excess of 30% m\(^{-1}\) is obtained for a linear current density of 250 mA cm\(^{-1}\). It was shown that, for a given discharge current, the optimum output power and gain of the laser are obtained for a discharge length for which the linear current density is approximately 65 mA cm\(^{-1}\). A threshold current of 280 mA was obtained for a 5 cm active length and bore diameter 4 mm with external mirrors each of transmission of 0.1%. This is an order of magnitude less than that achieved previously using a HC of the same active length and mirrors of the same transmission [5]. Both the threshold current and the threshold power are significantly lower than the respective values for a conventional hollow-cathode laser and show a markedly different dependence on the active length (23). It was also shown that the round-trip cavity loss for the SHC, due to the Brewster windows was approximately 2%. The fluorescence of the Brewster windows due to the laser beam is plainly visible.

An estimate of the CW beam power and the discharge power required for operation of the SHC with internal mirrors can be made using the following expression for the output power from a low-loss, homogeneously broadened, standing-wave laser [24]

$$P_{\text{out}} = 0.5 P_{\text{a}} T_{2} \left(2g_{0} \ell/(L + T_{2}) - 1\right).$$

(2)

The small-signal unsaturated gain per unit length is \(g_{0}\), the transmission of the output coupler is \(T_{2}\), the round-trip cavity loss (excluding \(T_{2}\)) is \(L\) and \(P_{a}\) is a saturation parameter.

In what follows it is assumed that the cross-sectional area of the beam remains constant. A multimode peak beam power of 0.7 mW was obtained from the above SHC for a current and voltage of 1 A and 1 kV, respectively. This gives an estimate of 10 mW for \(P_{a}\). If the laser is operated with internal mirrors so that \(L\) is reduced to 0.1%, then for \(T_{2} = 0.25\%\), an output power of 0.7 mW is obtained for an input power of 70 W (150 mA at 500 V). The current density is \(\sim 50\) mA cm\(^{-2}\). If the laser is repetitively pulsed (\(\sim 1000\) p/s) with a duty cycle of 0.1, the mean input power is reduced to 7 W. These calculations indicate that, even for an unoptimized bore geometry (diameter and cathode–anode area ratio), the laser could provide a useful ultraviolet beam for an instrument under conditions which reduce the cathode erosion and which are conducive to achieving a practical tube lifetime.

V. Ne–Cu\(^{+}\) 270-nm LASER

The dependence of the small-signal gain on the current for this transition (5s\(^{2}\)D\(_{1} \rightarrow 4p\(^{2}\)D\(_{1}\)) is shown in Fig. 4. This result forms part of a extended study of this laser currently in progress which will be reported elsewhere. The highest gain achieved is 12% m\(^{-1}\) at a linear current density of about 400 mA cm\(^{-1}\), which compares very favorably with the values of 3.6 [4] and 7 (combined lines) [25] % m\(^{-1}\) obtained by other workers for HC lasers using much higher current densities of 1.3 and 0.83 A m\(^{-1}\) respectively. The ratio of the small-signal gain per unit length to the linear current density is a maximum at a linear current density of about 220 mA cm\(^{-1}\). The threshold current for a 50 cm gain length was 6 A. The transmission of both laser mirrors was 0.3%. These results indicate that the value of \(L\) is about 2.8%. This emphasizes the need to operate the laser with internal mirrors. A peak beam power of 8 mW was obtained from each end of the laser for a current of 14 A. The single-pass gain at that current is about 5%. The application of (1) to this data gives a value of 24 mW for \(P_{a}\).

A projection of the performance with internal mirrors is made as follows. Assume an active length of 10 cm, a
current of 1 A, a value of 0.1 for \( L \) and an output coupler transmission of 0.2. Then, using (1), the pulsed output power is approximately 1.7 mW. The input power required for CW operation is just less than 500 W (1 A at 500 V), which is high. The corresponding current density is about 160 mA cm\(^{-2}\). However, continuously pulsed operation at a duty cycle of 0.01 would require a mean electrical input power of 5 W. It is stressed that the above results have been obtained using an unoptimized bore geometry.

VI. He–Cu\(^+\) VUV (150–160 nm) TRANSITIONS

As noted earlier, the He–Cu\(^+\) 6s → 4p transitions (150–160 nm) have a common upper level with the high gain infrared line (781 nm). For this reason the optimization of the gain on the 781-nm line is particularly relevant toward the demonstration of CW laser action on one or more of the VUV transitions. Eichler et al. [26] used emission spectroscopy to estimate the population inversion for each of the 6s → 4p transitions in a conventional slotted hollow-cathode discharge. The highest gain inferred from their results was 1.2 and 55% m\(^{-1}\) for the 154- and 781-nm lines, respectively, at a current density of 400 mA cm\(^{-2}\). They estimated that a gain as high as 5% per pass could be achieved using a 180-cm active length at a current density of 800 mA cm\(^{-2}\). Measurement of the gain by the above method is indirect and the result is sensitive to the value of the rate coefficient for spontaneous emission, a parameter that is subject to significant uncertainty. A projection based on our gain measurements for a 2-mm bore SHC tube [22] indicates that a gain of 110% m\(^{-1}\) on the 781-nm line could be obtained for a current density of 400 mA cm\(^{-2}\). On the basis of the measurements by Eichler et al., this would imply a gain of about 2.4% m\(^{-1}\) for the 154-nm line. However, we have observed that the intensity of the spontaneous emission on the 154.17-nm line from a 3-mm bore tube is about four times that obtained from the same tube when operated as a HC (27) at the same electrical input power. This direct comparison suggests that the attainable gain at that wavelength for the SHC may be as much as four times the above value estimated for a HC by Eichler et al. A more detailed examination of the Cu\(^+\) spectrum at 150–160 nm is in progress and will be reported elsewhere. Our gain measurements for the SHC on the He–Au\(^+\) 282 and the Ne–Cu\(^+\) 270-nm ultraviolet lines show that the gain (% m\(^{-1}\)) is significantly greater than that reported for these lines using a conventional hollow-cathode. This superior UV performance of the SHC tube also suggests that it may also give a significantly higher specific gain in the VUV than that estimated for an HC tube by Eichler et al. Such higher gain is critical for the successful demonstration of proposed VUV laser since the currently attainable mirror reflectance at the above VUV wavelengths is between 92 and 96%. For this reason the single-pass gain must be at least 8% m\(^{-1}\). Furthermore, the best VUV window material at these wavelengths, MgF\(_2\), has a transmission of 80–85% at 150 nm for a thickness of 2.5 mm so that internal mirrors must be employed. Consequently, the development of a bakeable form of the SHC tube with internal mirrors is considered to be an essential step for demonstrating the VUV laser.

VII. CONCLUSION

The segmented hollow-cathode pumping discharge gives a higher gain on the He–Cu\(^+\) 781, He–Au\(^+\) 282, and Ne–Cu\(^+\) 270-nm lines at lower discharge current and input power than do the conventional-cathode, hollow anode cathode and helical hollow-cathode discharges. In particular, it has been shown that a He–Au\(^+\) 282-nm laser with internal mirrors operating at a pulse repetition rate of about 1000 p/s could generate a peak power of the order of 0.5 mW for an average electrical input power of 7 W. Thus the segmented hollow-cathode offers the prospect of compact low-cost repetitively pulsed ultraviolet lasers for use as economic light sources for instruments in biotechnology. The projections for increasing the gain on the He–Cu\(^+\) VUV lines indicate that laser action on one or more of these lines may be achieved.

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REFERENCES


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