# Helium 2<sup>3</sup>S Densities and Excitation Mechanisms in a Hollow Cathode He–Cd<sup>+</sup> Laser

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#### Abstract

Helium singlet and triplet metastable number densities have been measured in the active region of a hollow cathode He–Cd<sup>+</sup> laser. The results are shown to be consistent with measurements of the electron density and energy distribution functions obtained in other experiments. The excitation mechanisms for the 441.6 nm Cd<sup>+</sup> laser transition have been investigated by comparing the parametric behaviour of the triplet metastable number density, the 441.6 nm spontaneous emission and the laser output power. Unlike positive column He–Cd<sup>+</sup> lasers, Penning ionization is found to be only partially responsible for populating the upper laser level. Electron impact excitation is shown to be the major pumping mechanism for both the upper and lower laser levels, and it is the faster rate of this process for the lower level which is responsible for the observed saturation of laser power with increasing discharge current.

# **1. Introduction**

Helium-cadmium lasers operating in the positive column region of a gas discharge have been the subject of many investigations aimed at understanding the excitation mechanisms and optimizing the performance (see e.g. Browne and Dunn 1973; Miyazaki et al. 1974; Johnston and Kolb 1976; Mori et al. 1978). However, metal vapour lasers operating in the negative glow region of the discharge have not been subjected to the same degree of investigation. The general excitation processes in these hollow cathode lasers have been studied and the reasons for the negative glow region of the discharge being an especially favourable one for the excitation of nearly all CW metal ion laser transitions have been established (Gill and Webb 1977a, 1977b). Some specific systems have been examined in varying degrees of detail (He-I<sup>+</sup>, Shay et al. 1975; He-Hg<sup>+</sup>, Kano et al. 1975; He-Zn<sup>+</sup>, Gill and Webb 1978; He-As<sup>+</sup>, Piper et al. 1978), but, apart from reports of laser oscillation and of parametric variations in output power (Piper and Webb 1973; Fujii 1973; Fujii et al. 1975; Csillag et al. 1977; Hernqvist 1978; Grace and McIntosh 1979), the hollow cathode He-Cd<sup>+</sup> system has not been the subject of detailed diagnostic experiments. This omission is surprising because there are a number of reasons why the hollow cathode He-Cd<sup>+</sup> laser is an attractive candidate for further development. Firstly, like the conventional positive column He-Cd<sup>+</sup> laser, it offers outputs at blue (441.6 nm) and ultraviolet (325.0 nm) wavelengths but, because of differences between the two types of discharge, the output power of the hollow cathode laser at these wavelengths is not necessarily subject to the same limitations as the positive column device. Secondly, the high frequency output noise which is troublesome in conventional 441.6 nm Cd<sup>+</sup>

lasers is greatly reduced in the hollow cathode configuration because the striation waves which modulate the output are prevented from growing to significant amplitudes by the very small anode-cathode spacing (Willgoss and Thomas 1974; Johnston and Kolb 1976). Finally, the fact that simultaneous oscillation can be achieved at red ( $635 \cdot 5$ ,  $636 \cdot 0$  nm), green ( $533 \cdot 7$ ,  $537 \cdot 8$  nm) and blue ( $441 \cdot 6$  nm) wavelengths makes this system a possible candidate for development as a white light laser (Fujii *et al.* 1975).

In positive column He–Cd<sup>+</sup> lasers it is clearly established (Janossy *et al.* 1972; Browne and Dunn 1973) that the dominant mechanism for populating the upper level of the  $441 \cdot 6$  nm laser transition is the Penning reaction

$$He(2^{3}S) + Cd \rightarrow (Cd^{+})^{*} + He + e.$$
 (1)

Although there is some evidence that electron impact excitation may be significant (Mori *et al.* 1978), it is generally considered that saturation of the laser output with increasing discharge current is due to saturation of the population of the He( $2^3S$ ) pump species. These metastables are created by electron impact excitation of ground state helium atoms and are lost principally by electron impact ionization (Browne and Dunn 1973). Consequently, in a positive column discharge for which the electron energy distribution is Maxwellian and changes little with current, and for which the electron density rises with increasing current, the population of metastables ceases to rise when the electron density reaches a certain level.

By contrast, the electron energy distribution in a hollow cathode discharge is very highly non-Maxwellian and varies dramatically with current, pressure and radial position (Gill and Webb 1977*a*, 1977*b*) and it is by no means clear that the Penning reaction will assume the same relative importance in these circumstances. McIntosh *et al.* (1978) did not observe saturation of the He( $2^3S$ ) density over the range of currents investigated in a pure helium hollow cathode discharge, while in the case of He–Zn Gill and Webb (1978) showed that electron impact excitation may account for up to 70% of the upper level population of the Zn<sup>+</sup> laser transition at 589 4 nm which was previously thought to be excited almost entirely by Penning ionization.

Grace and McIntosh (1979) have reported a design for a hollow cathode He–Cd<sup>+</sup> laser with ouput powers exceeding 100 mW at each of  $441 \cdot 6$  and  $533 \cdot 7-537 \cdot 8$  nm and with low noise and good long-term stability. This laser, in common with other devices of the same general type, has output at  $441 \cdot 6$  nm which depends markedly on the cadmium concentration, the helium pressure and the gas flow rate and which ultimately saturates with increasing discharge current. By measuring the He(2<sup>3</sup>S) density as a function of these same discharge parameters it is possible to assess the relative importance of the Penning reaction of equation (1).

#### 2. Discharge Tube

The hollow cathode discharge tube was the same as that described by Grace and McIntosh (1979). The cathode consisted of a long stainless steel tube (length 60 cm, bore 4 mm) with 14 separately ballasted anodes distributed along its length. Helium gas was introduced at the tube centre and flowed along to the ends of the tube where it was exhausted by a backing pump. The gas pressure was monitored at the inlet. Metal vapour was introduced at varying rates by controlling the temperature of an oven placed in a side arm at the tube centre.

# 3. Measurement of Metastable Helium Population Densities

The long lifetimes of the metastable states dictate the use of an absorption technique to measure their population densities. Of the various possible techniques of line absorption, those of Harrison (1959) and McConkey (1969) are conveniently applied to the measurement of excited state densities in a gas discharge since for neither of these is it necessary to achieve the difficult task of maintaining two separate but identical discharges. The method described by McConkey, in which the light signals emitted from a known volume of the discharge are measured as the length of the active discharge is increased, is easily applied to the hollow cathode configuration because switching the individual anodes enables the length of the discharge to be varied in a known manner.

With a system of apertures used to ensure that each volume element along the axis of the discharge radiates the same amount of light into a spectrometer, the ratio of intensities  $I_1$  and  $I_2$  for discharges of lengths  $d_1$  and  $d_2$  is given by

$$\frac{I_1}{I_2} = \frac{\int_0^\infty \{1 - \exp(-k_0 d_1 e^{-\omega^2})\} d\omega}{\int_0^\infty \{1 - \exp(-k_0 d_2 e^{-\omega^2})\} d\omega}$$
(2)

(McConkey 1969), in which the value of the absorption coefficient  $k_0$  to be deduced is related to the population density by the standard relationship of Mitchell and Zemansky (1961).

As adapted to the present experiment, the intensity ratio of equation (2) was measured by comparing the intensity of the light emitted by a discharge with a single anode with that emitted by a discharge with a number of anodes chosen so that their negative glows did not overlap. The length  $d_1$  was thus an integral multiple of  $d_2$ , the values of the latter being taken from the results of Grace and McIntosh (1979) who measured the length of discharge associated with a single anode for various discharge conditions in the same experimental tube. The large axial extent of the negative glow associated with a single anode when the discharge contained  $\sim 0.1$  torr (13.3 Pa) of cadmium, together with the requirement that the multiple glows should not overlap and therefore interact, made it necessary to restrict the ratio of discharge lengths to 3:1 or less and this consequently limited the accuracy with which low densities of metastables could be measured. It was not possible to overcome this restriction by the use of a longer discharge tube because the collection angle for light from the far end of the tube was approaching the diffraction limit for even the present length of 60 cm.

The He( $2^{1}$ S) and He( $2^{3}$ S) populations were determined from measurements of absorption at 501 · 6 and 388 · 9 nm respectively. Allowance was made for the hyperfine splitting of the triplet transition at 388 · 9 nm in the manner described by Browne and Dunn (1973). The calculated intensity ratio is effectively independent of gas temperature over the range of likely temperatures, but a gas temperature of 620 K, equal to the wall temperature of the discharge tube (Grace and McIntosh 1979), was used to calculate the Doppler widths of both lines.

In deducing populations from the measured absorption it is necessary to assume that the populations of the upper levels of the monitored transitions are negligible. This may be checked for the 2<sup>1</sup>S population by measuring the ratio of absorption coefficients for the 501.6 and 396.5 nm transitions, both of which terminate on the metastable level; in the absence of significant upper level populations the absorption coefficients should be in the ratio  $\lambda_1^3 A_1 : \lambda_2^3 A_2$ , where  $\lambda_1$  and  $\lambda_2$  are the wavelengths of the transitions and  $A_1$  and  $A_2$  the corresponding Einstein coefficients, the value of the ratio being 3.78:1 for the present case (Browne and Dunn 1973). No systematic deviation from a value of  $3.6\pm0.5$  was observed in the present experiments, the large uncertainty being due to the weak signals obtained at 396.5 nm. There was insufficient intensity at any line other than 388.9 nm terminating on the 2<sup>3</sup>S level to make the corresponding check for the triplet metastable populations. In view of the fact that the observed ratio was close to that expected in the absence of upper level effects, and in the absence of any further information, it was assumed that the populations of the upper levels were negligible and hence all intensity ratio measurements were converted directly to singlet and triplet metastable populations.

Radial variations in the populations of the emitting and absorbing species in hollow cathode discharges have been observed by Gill and Webb (1978) and McIntosh *et al.* (1978). Since the absorption experiment requires the assumption of a homogeneous discharge, the effect of these radial variations was made negligible by restricting the width of the cone along the axis from which light was accepted by the spectrometer. The efficacy of this was tested in two ways: (1) by routinely checking that the signal collected by the optical system from each separate anode was the same (to within  $\pm 10\%$ ) and exhibited no systematic variation along the length of the tube, and (2) by checking that the measured absorption coefficient was independent of the anodes used in its determination and thus of the distance from the spectrometer to the active region of the discharge.

The unavoidable flaring of the negative glow associated with each anode limited the accuracy with which absolute metastable populations could be measured to  $\pm 20\%$ , and other effects may further increase the uncertainty. However, data taken under nominally identical conditions on different occasions varied by only 5–10% and it is considered that the relative populations were determined with ample accuracy to justify the conclusions reached.

# 4. Results and Discussion

### (a) Metastable Densities

The  $2^{3}$ S and  $2^{1}$ S helium metastables, with thresholds of  $19 \cdot 8$  and  $20 \cdot 6$  eV respectively, the  $3^{3}$ P level which is the upper level for the  $388 \cdot 9$  nm emission (threshold 23 eV) and the 468  $\cdot 6$  nm He II emission (threshold 75 eV) are all produced by the high energy electrons in the discharge. As shown in Fig. 1 the spontaneous emission at 388  $\cdot 9$  nm falls only slightly as the pressure of cadmium in the discharge is increased from essentially zero at 200°C to  $0 \cdot 1$  torr at 320°C. The spontaneous emission data have been corrected for the effects of self-absorption in the manner described by Webb (1964). The fact that the 468  $\cdot 6$  nm He II emission behaves in an almost identical fashion to the 388  $\cdot 9$  nm emission shows that it is the high energy electrons that are responsible for the  $3^{3}$ P excitation and not the low energy electrons through stepwise excitation from the  $2^{3}$ S level. Since the spontaneous emission declines by only a factor of 4 and this emission arises from the impact of electrons of high energy, we can conclude that the production rate of helium metastables does not suffer a reduction

of more than a factor of 4 as the cadmium concentration increases through the range of vapour pressures of interest. Over the same range of cadmium concentrations, the axial population density of  $2^{3}$ S metastables is shown by Fig. 1 to decline by over two orders of magnitude. Since the production rate is sensibly constant while the equilibrium density falls, it is clear that the loss rate for the  $2^{3}$ S metastables must be increasing as the proportion of cadmium in the discharge rises. With the current and pressure constant, the only mechanisms which will result in increasing loss of  $2^{3}$ S metastables are the Penning reaction of equation (1) or a change in the electron energy distribution leading to increased losses from the metastable state.



Fig. 1. Variation of helium triplet metastable number density  $N(2^{3}S)$  and 388.9 nm spontaneous emission with changes in oven temperature *T*. The data are for the experimental conditions: helium pressure p = 20 torr, discharge current  $I_{D} = 150 \text{ mA}$  per anode, gas flow rate  $G = 100 \text{ atm cm}^{3} \text{min}^{-1}$ . (Note 1 torr  $\equiv 133 \text{ Pa}$ ; 1 atm  $\equiv 101 \text{ kPa}$ .)

The variation with discharge current of the triplet and singlet metastable helium densities is shown in Fig. 2 for a selection of gas flow rates at a fixed oven temperature and helium pressure. Since the proportion of cadmium in the discharge decreases as the flow rate increases at a fixed oven temperature, the variations of  $2^3$ S density with flow rate in this diagram are consistent with the data for changes in oven temperature described by Fig. 1. The saturation of metastable densities with increasing current is due to the balance between the production by electron impact excitation from ground state atoms and the losses by electron impact and Penning ionization. Electron impact ionization is the most important loss mechanism, since, compared with the positive column discharge for which a similar conclusion was reached by Browne and

Dunn (1973), the loss of metastables by diffusion is lower (because of the higher pressure), the loss by metastable-metastable collisions is lower (because of the lower metastable density), the loss by metastable-ground state collisions increases by a factor ~5 but remains negligible, while the loss by electron impact ionization is higher (because of the much higher electron densities in the hollow cathode discharge; Belal and Dunn 1978). The singlet densities were between 0.2 and 0.3 of the triplet density and generally varied in the same manner as the triplet densities with changes in the discharge conditions. The  $2^{3}$ S densities and their dependence on discharge current shown in Fig. 2 (full curves) are similar to those obtained in helium-zinc by Gill and Webb (1977*a*) and are generally similar in shape but about a factor of 2 less than the values obtained in a helium discharge by McIntosh *et al.* (1978).



Fig. 2. Variation of helium singlet and triplet metastable number densities  $N_{\rm m}$  with discharge current  $I_{\rm D}$  for several gas flow rates G. Conditions: p = 20 torr,  $T = 320^{\circ}$ C.

Although Browne and Dunn (1973) found that Penning ionization was not a significant loss process for  $2^{3}$ S metastables in a positive column discharge, this does not appear to be the case in the hollow cathode device. The relative loss f due to Penning collisions and electron impact ionization is

$$f = N_{\rm Cd} \langle \sigma_{\rm P} V \rangle / n_{\rm e} \langle q_{\rm i} v \rangle,$$

in which  $N_{Cd}$  is the cadmium number density,  $\sigma_P$  is the cross section for Penning ionization, V is the relative velocity of helium and cadmium atoms,  $n_e$  is the electron density,  $q_i$  is the cross section for ionization of the metastables and v is the electron velocity. We can approximate the average term in the numerator as  $\bar{\sigma}_P \bar{V}$ , where  $\bar{\sigma}_P$ is taken from the data of Riseberg and Shearer (1971) and  $\bar{V} = (8kT/\pi\mu)^{\frac{1}{2}}$  for a Maxwellian distribution. Calculation of the denominator is more difficult since we have no precise data for either  $n_e$  or the electron energy distribution function. However, in a 3 mm bore He–Zn discharge Gill and Webb (1978) found  $n_e \sim 6 \times 10^{13}$  cm<sup>-3</sup>, and in a 6 mm bore He discharge Belal and Dunn (1978) found  $n_e \sim 2 \times 10^{13}$  cm<sup>-3</sup>, both values being for currents of 150 mA per anode and a pressure of 20 torr. Hence a value  $n_e \sim 4 \times 10^{13}$  cm<sup>-3</sup> should not be significantly in error for the present conditions. There have been no complete determinations of the electron energy distribution but Gill and Webb (1977*a*) have outlined the general form that the distribution takes and, as a crude approximation, we can construct a function which exhibits the features they describe. The data of Long and Geballe (1970), suitably extrapolated to high energies, were used for  $q_i$ . The conclusion reached is only weakly dependent on the actual form of these approximations provided the energy distribution does contain a high energy tail extending out to the cathode fall voltage



Fig. 3. Variation of helium triplet metastable number density  $N(2^{3}S)$  with helium pressure p for the two indicated gas flow rates G. Conditions:  $I_{\rm D} = 150$  mA per anode,  $T = 320^{\circ}C$ .

of the discharge as well as a peak at  $\sim 3 \text{ eV}$  containing most of the electrons. In this way we estimate that the loss of  $2^3$ S metastables due to the Penning reaction may be as much as 50% of that due to electron impact ionization.

The variation of  $2^{3}$ S densities with helium pressure is shown in Fig. 3. The  $2^{3}$ S densities are higher at an increased flow rate; this change is due to the change with flow rate of the proportion of cadmium in the discharge and is thus consistent with the data of Figs 1 and 2.

If electron impact excitation is the only creation process and electron impact and Penning ionization are the only significant loss processes for the metastables, the curves of Fig. 3 will be described by an equation of the form

$$N_{\rm m} = \langle q_{\rm e} v \rangle n_{\rm e} N_0 / (\langle q_{\rm i} v \rangle n_{\rm e} + \langle \sigma_{\rm P} V \rangle N_{\rm Cd}).$$
<sup>(3)</sup>

Here  $N_0$  is the ground state helium density,  $q_e$  is the cross section for excitation of the 2<sup>3</sup>S level, and the other symbols are as defined above. To evaluate equation (3) we require information about the electron density  $n_e$  and the electron energy distribution function  $f(\varepsilon)$  as functions of the discharge parameters. This information is not available for the specific geometry or system being investigated and hence a quantitative study is not possible. However, there is general agreement about the way in which  $n_e$  and  $f(\varepsilon)$  vary in related experiments and we may use this information to explain qualitatively the variation of the metastable density with pressure.

Belal and Dunn (1978) found that the electron density in a 6 mm bore hollow cathode helium discharge rose to a maximum of  $5 \times 10^{13}$  cm<sup>-3</sup> at 10 torr and then decreased again as the pressure was increased. Gill and Webb (1978) obtained similar results in a 3 mm bore helium-zinc tube, with the maximum  $n_e$  being  $7 \times 10^{13}$  cm<sup>-3</sup> at 30 torr. A similar functional form may be assumed for the present 4 mm bore helium-cadmium discharge, with the peak electron density being expected at around 20 torr. Since the data of Fig. 3 are for a current of 150 mA per anode which, by Fig. 2, is known to be a current at which the metastable population is saturated with respect to changes in electron density, the slow increase in  $N_m$  for pressures greater than 25 torr may be partly accounted for by the decline in axial values of  $n_e$ . However, both  $n_e$  and  $N_0$ rise for pressures less than 20 torr and another explanation must be sought for the sharp fall in  $N_m$ .

Electron energy distribution functions  $f(\varepsilon)$  have been measured in hollow cathode helium discharges of varying tube diameter, pressure and current density by Borodin and Kagan (1966), Soldatov (1971) and Kagan and Taroyan (1973). Although differing substantially in many respects, the results of all these experiments do show that the most probable value of the electron energy falls as the pressure is increased. Similar conclusions were reached by Gill and Webb (1977a), whose data show that the most probable energy of the 'plasma component' (containing most of the electrons) decreases rapidly over the pressure range of the present experiments. The threshold for  $q_e$  is 19.8 eV while that for  $q_i$  is ~5 eV and consequently any shift to lower energies of the plasma component of the energy distribution will cause an increase in  $\langle q_i v \rangle$  and a smaller decrease in  $\langle q_e v \rangle$  which depends rather more on the 'beam' electrons that are less affected by the change in pressure. Although the lack of quantitative data prevents any calculation of  $N_{\rm m}$  for comparison with the experimental data, changes of  $\langle q_e v \rangle$ ,  $\langle q_i v \rangle$  and  $n_e$  in the manner described do account qualitatively for the observed variation of the 2<sup>3</sup>S density with pressure.

# (b) Excitation Mechanisms for the 441.6 nm Laser Transition

Hollow cathode He–Cd<sup>+</sup> lasers operating at 441.6 nm are characterized by an optimum oven temperature, an optimum gas pressure, an optimum gas flow rate and and an output power level which saturates with increasing discharge current. If the Penning reaction of equation (1) is solely responsible for creating the population inversion, the 441.6 nm output would be expected to correlate with the product  $N_m N_{Cd}$ .

It was not possible to measure  $N_{Cd}$  by absorption in the present discharge tube and therefore the value calculated from the vapour pressure set by the oven temperature was used as an approximation. Although the cadmium number density in the tube will not be equal to this value, it should be directly proportional to it if the gas flow rate and cathode temperature are unchanged. There may be some dependence of  $N_{\rm Cd}$  on discharge current but the fact that the intensity of Cd<sup>+</sup> lines excited by charge transfer varies linearly with current up to the maximum current used (see e.g. Piper and Webb 1973) shows that there is ample cadmium present for  $N_{\rm Cd}$  not to be a limiting factor. This will be even more true for the Penning excited lines because the metastable densities are about two orders of magnitude less than the helium ion densities.



**Fig. 4.** Variation with oven temperature *T* of the helium triplet metastable number density  $N(2^{3}S)$ , the saturated cadmium partial pressure  $p_{cd}$ , the 441  $\cdot$ 6 nm output and the product of the triplet metastable and cadmium number densities  $N(2^{3}S) N_{cd}$ . The vertical scales for the 441  $\cdot$ 6 nm output and  $N(2^{3}S) N_{cd}$  are arbitrary. Conditions: p = 20 torr,  $I_{D} = 150$  mA per anode, G = 100 atm cm<sup>3</sup> min<sup>-1</sup>.

When the gas flow rate is increased at a fixed oven temperature, the cadmium density in the tube decreases while, as shown in Figs 2 and 3, the 2<sup>3</sup>S density increases. Hence, as noted by Grace and McIntosh (1979), the existence of an optimum flow rate is readily explained. More direct comparisons between  $N_m N_{Cd}$  and the 441  $\cdot$  6 nm output must be restricted to circumstances in which the gas flow rate and cathode temperature are not changed. Fig. 4 shows the results of such a comparison when the oven temperature is varied. The absolute scale for the laser power is of no significance but the similarity in shape between this curve and that for  $N_m N_{Cd}$  is strong evidence that Penning ionization is at least partly responsible for creating the population inversion. There is, however, some evidence in Fig. 4 that other excitation mechanisms are important, and this is brought out more clearly in Fig. 5, which shows a comparison

between the variations with current of the  $2^{3}S$  density and the  $441 \cdot 6$  nm spontaneous emission. An even more striking demonstration of the importance of these other processes is given in Fig. 6, which shows that the variation of  $441 \cdot 6$  nm laser output with pressure is quite unlike that of the  $2^{3}S$  density.

Although the metastable density saturates with increasing discharge current in Fig. 5, the 441.6 nm spontaneous emission does not but in fact rises linearly with current beyond the value at which metastable saturation occurs. Some contribution to this increase may come from Penning collisions with non-metastable atoms but, as shown in Section 3, populations of these levels were small compared with those



Fig. 5. Comparison between helium triplet metastable number density  $N(2^{3}S)$  and 441.6 nm spontaneous emission as functions of the discharge current  $I_{\rm D}$ . Conditions: p = 20 torr,  $T = 320^{\circ}C$ , G = 50 atm cm<sup>3</sup> min<sup>-1</sup>.

of the metastable levels. Even if the Penning cross section for these levels is assumed equal to that of the metastable level, it is not possible to account for the observed 441.6 nm emission. On the other hand, the electron density increases linearly with current (Gill and Webb 1978; Belal and Dunn 1978) and it is electron impact excitation that is responsible for the continued increase in the population of the  $5s^{2} {}^{2}D_{5/2}$  cadmium ion level.

Janossy *et al.* (1972) argued that electron collisions with ground state  $Cd^+$  ions were important and assumed that the ions were produced by the Penning process. More recently, Mori *et al.* (1978) have shown that this assumption cannot be correct and they demonstrate instead that, in a positive column discharge, two stages of

electron impact excitation occur, one from the atomic to the ionic ground state and the other upwards from the ionic ground state. They estimate that the rate for this two-step process is an order of magnitude greater than that for direct excitation from the atomic ground state to an excited ionic state. This is not true for a hollow cathode discharge in which significant numbers of electrons have energies in the range for which the cross section for direct excitation reaches the unexpectedly large value of  $1.5 \times 10^{-16}$  cm<sup>2</sup> (Aleinikov and Ushakov 1970); indeed a calculation based on an energy distribution of the form described by Gill and Webb (1977*a*), the cross section of Aleinikov and Ushakov for direct excitation and that given by the formula



Fig. 6. Comparison between triplet metastable number density  $N(2^{3}S)$  and 441.6 nm multimode laser power  $P_{441.6}$  as functions of the helium pressure p. Conditions:  $I_{D} = 150$  mA per anode,  $T = 320^{\circ}C$ , G = 50 atm cm<sup>3</sup> min<sup>-1</sup>.

of Blaha (1969) for stepwise excitation, suggests that the direct process is likely to be the more important in the hollow cathode case. The calculated rate for electron impact excitation of the  $5s^{2} {}^{2}D_{5/2}$  level is several times larger than the rate due to Penning collisions and this agrees with the conclusion of Gill and Webb (1978) that 70% of the excitation of the upper level of the 589.4 nm Zn<sup>+</sup> transition was due to electron collisions and only 30% to the Penning process.

Although the population of the upper level does continue to rise with increasing current, Fig. 7 shows that the 441.6 nm laser power saturates for total discharge currents of around 2.5 A in the present tube. This indicates that lower level population effects must become important as the discharge current (and hence  $n_e$ ) rises. The

cross section for Penning excitation of the lower level is 0.38 that of the upper level (Cermak 1971) and the rate for this process will follow the same trends with current as that for the upper level. The cross section for direct electron impact excitation of the lower level is  $\sim \frac{1}{3}$  that of the upper level (Varshavskii *et al.* 1970) and this is only very slightly offset by the fractionally lower threshhold energy. On the other hand, the cross section for stepwise excitation from the Cd<sup>+</sup> ground state to the lower laser level has a significantly lower threshhold (5.8 eV to the lower level and 8.6 eV to the upper level) and corresponds to an optically allowed transition with a much larger cross section. Thus Penning ionization will make only a small contribution to the population of the lower level whereas the contribution from electron impact will be larger than it is for the upper state. Hence as the discharge current and electron density rise beyond the point at which the metastable density saturates, the population of the lower level increases at a faster rate than that of the upper level and the laser output power saturates, as shown in Fig. 7.



Fig. 7. Variation of 441 6 nm TEM<sub>00</sub> laser power  $P_{441.6}$  with discharge current  $I_{\rm D}$ . Conditions: p = 20 torr, T = 320°C, G = 70 atm cm<sup>3</sup> min<sup>-1</sup>.

The 441.6 nm laser power peaks at around 20 torr in the present tube (see Fig. 6), at which pressure the metastable density is falling rapidly while, based on the results of Gill and Webb (1978) and Belal and Dunn (1978), the axial electron density is expected to have its maximum value. This again suggests that electron impact excitation plays a major role in populating the laser levels. A fuller explanation requires rate calculations which incorporate changes with pressure of both the electron density and the electron energy distribution. Such calculations, together with others for the situations in which estimates have been given above, are now being made and ultimately will be reported elsewhere.

### 5. Conclusions

Helium singlet and triplet metastable number densities have been measured in the active region of a hollow cathode He-Cd<sup>+</sup> laser. In the absence of precise knowledge of the electron density  $n_e$  and energy distribution function  $f(\varepsilon)$ , it was not possible to calculate the expected metastable densities for comparison with the experimental values. However, estimates based on the knowledge of  $n_e$  and  $f(\varepsilon)$  gained in related experiments do account for the observed variations in metastable populations.

The excitation mechanisms for the  $441 \cdot 6$  nm Cd<sup>+</sup> laser were investigated by comparing the parametric behaviour of the triplet metastable number density and the laser output power. Unlike the situation in positive column lasers, Penning ionization was found to be only partially responsible for populating the upper level of the laser transition. The greater numbers and higher energies of the electrons in a hollow cathode discharge increase the importance of electron impact excitation and this appears to be the major pumping mechanism. Further, electron impact excitation of the lower laser level is also important and it is the faster rate of this process which is responsible for the observed saturation in output power with increasing discharge current.

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#### References

Aleinikov, V. S., and Ushakov, V. V. (1970). Opt. Spectrosc. (USSR) 29, 111-12.

- Belal, I. K., and Dunn, M. H. (1978). J. Phys. D 11, 303-23.
- Blaha, M. (1969). Astrophys. J. 157, 473-7.
- Borodin, V. S., and Kagan, Y. M. (1966). Sov. Phys. Tech. Phys. 11, 131-4.
- Browne, P. G., and Dunn, M. H. (1973). J. Phys. B 6, 1103-17.
- Cermak, V. (1971). Collect. Czech. Chem. Commun. 36, 948-50.
- Csillag, L., Nam, C. Z., Janossy, M., and Rozsa, K. (1977). Opt. Commun. 21, 39-41.
- Fujii, K. (1973). Electr. Eng. Jpn 93, 106-14.
- Fujii, K., Takahashi, J., and Asami, Y. (1975). IEEE J. Quantum Electron. QE 11, 111-14.

Gill, P., and Webb, C. E. (1977a). J. Phys. D 10, 299-311.

- Gill, P., and Webb, C. E. (1977b). J. Phys. D 10, 2235-44.
- Gill, P., and Webb, C. E. (1978). J. Phys. D 11, 245-54.
- Grace, J. R., and McIntosh, A. I. (1979). Design and performance of an improved hollow cathode He-Cd<sup>+</sup> laser. J. Phys. D (in press).

Harrison, J. A. (1959). Proc. Phys. Soc. London 73, 841-8.

Hernqvist, K. G. (1978). IEEE J. Quantum Electron. QE 14, 129-32.

Janossy, M., Itagi, V. V., and Csillag, L. (1972). Acta. Phys. Acad. Sci. Hung. 32, 149-63.

Johnston, T. F., and Kolb, W. P. (1976). IEEE J. Quantum Electron. QE 12, 482-93.

Kagan, Y. M., and Taroyan, A. S. (1973). Opt. Spectrosc. (USSR) 35, 120-3.

Kano, H., Shay, T., and Collins, G. J. (1975). Appl. Phys. Lett. 11, 610-12.

Long, D. R., and Geballe, R. (1970). Phys. Rev. A 1, 260-5.

McConkey, J. W. (1969). J. Opt. Soc. Am. 59, 1262-6.

McIntosh, A. I., Dunn, M. H., and Belal, J. K. (1978). J. Phys. D 11, 301-11.

Mitchell, A. G., and Zemansky, M. W. (1961). 'Resonance Radiation and Excited Atoms' (Cambridge Univ. Press).

Miyazaki, K., Ogata, Y., Fujimoto, T., and Fukuda, K. (1974). Jpn J. Appl. Phys. 13, 1866-74.

Mori, M., Murayama, M., Goto, T., and Hattori, S. (1978). IEEE J. Quantum Electron. QE 14, 427-33.

Piper, J. A., Littlewood, I. M., and Webb, C. E. (1978). J. Phys. B 11, 3731-44.

Piper, J. A., and Webb, C. E. (1973). J. Phys. D 6, 400-7.

Riseberg, L. A., and Shearer, L. D. (1971). Phys. Lett. A 35, 269-70.

- Shay, T., Kano, H., and Collins, G. J. (1975). Appl. Phys. Lett. 26, 531-4.
- Soldatov, A. N. (1971). Opt. Spectrosc. (USSR) 31, 97-101.

Varshavskii, S. P., Mityureva, A. A., and Penkin, N. P. (1970). Opt. Spectrosc. (USSR) 29, 341-3.

- Webb, C. E. (1964). D.Phil. Thesis, Oxford University.
- Willgoss, R. A., and Thomas, G. C. (1974). J. Phys. D 7, 2269-76.

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