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Laser Spectroscopy of a Pulsed Mercury-Helium Discharge

A. L. Bloom, W. E. Bell, and F. O. Lopez
Spectra-Physics, Inc., Mountain View, California

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Following the discovery of visible laser transitions in a pulsed mercury-helium discharge, a systematic search has been made for additional laser lines between the visible region and 2μ. A total of 19 lines has been observed, including the four listed by Bell and two reported earlier in a continuous mercury glow discharge. Regularities in the transitions which permit laser operation indicate that population inversions among Hg II levels arise from direct electron impact excitation from the Hg ground state, whereas inversions in Hg I more likely arise from indirect or cascade processes.

INTRODUCTION

THE possibility of laser action in mercury vapor discharges is of interest because of the relatively simple energy level structure and the ease with which mercury vapor can be handled in the laboratory. The first successful experiments were done in a continuous glow discharge[1,2] and revealed two laser transitions in the infrared, at around 1.5 and 1.8 μ. More recently, Bell[3] has found strong laser lines, including two in the visible, from the Hg II spectrum in a pulsed mercury-helium discharge. The present paper reports a continuation of this work consisting of a survey for lines in the infrared out to about 2 μ. A total of 19 lines are now known, including several additional ones in Hg I. Many of the new lines observed in Hg II have apparently not been reported previously in the literature.

The apparatus used for this work was a 15-mm bore laser and pulsing network described earlier,[3] but shortened from the original length of 3 m to about 2.25 m. A set of mirrors was prepared that provided 99% or better reflectance for every wavelength within the survey range. The pulse duration was about 3 μsec, with peak currents variable from 10 to 50 A, and the He pressure was varied from 0.8 to 1.2 Torr. The Hg partial pressure was approximately the vapor pressure at 45°C.

RESULTS AND DISCUSSION

A summary of the results is given in Table I. The wavelength measurements were made on a grating monochromator calibrated on the visible Hg and Ne laser lines, and it is felt that most of the discrepancies between observed wavelengths and assumed transitions[4] are within the probable systematic errors of the calibration. A possible exception is the line at 8628 Å, for which a fit to the nearest reasonable transition at 8622 Å is well outside any calibration error. The assignment of this transition should, therefore, be considered provisional, or it may represent an error in the energy determination of the relatively poorly excited lower level.

Four of the observed lines could not be matched to any possible transition between listed Hg II levels.[4] A correspondingly thorough search has not been made through the Hg I term values; however, all of the laser lines definitely assignable to Hg I are also listed in compilations of Hg spectrum lines.[5] The existence of Hg I laser lines in this part of the spectrum that had not previously been seen in emission seems possible but unlikely. More likely, these are Hg II transitions from highly excited $5d^{10}(^1S)$ states to unlisted states involving excited d electrons. If oscillator strengths permit, laser transitions of this character are quite probable, since the $5d^{10}(^1S)$ states are much more likely to be excited in direct electron collisions than are the others, and should result in many population inversions between highly excited levels.

As the data on the Hg II laser transitions accumulates, a number of interesting regularities are becoming

Table I. Observed laser transitions.

λ Measured	λ Theor.	Hg spectrum	Transition[a]	Reference
5678	5677	II	$5f_{7/2}-6d_{5/2}$	3
6150	6150	II	$7p_{3/2}-7s_{1/2}$	3
7346	7346	II	$7d_{5/2}-7p_{3/2}$	3
8547	8548	II	$5g-C$	
8628	8622	II	$8p_{3/2}-{}^4D^2{}_{5/2}$	
8677				
9396	9396	II	$10s_{1/2}-8p_{3/2}$	
10 586	10 583	II	$8s_{1/2}-7p_{3/2}$	3
11 181	11 179	II	$7g-6f_{5/2}$	
12 545				
12 981				
13 655				
15 288	15 295	I	$6p'3P_2-7^3S_2$	1, 2
15 550	15 554	II	$7p_{3/2}-6d_{5/2}$	
16 916	16 918	I	$5^1F_3-6^1D_2$	
	16 921	I	$7^3D_3-7^3P_2$	
16 939	16 942	I	$5^3F_2-6^3D_1$	
17 070	17 073	I	$7^1D_2-7^3P_2$	
17 112	17 110	I	$5^3F_3-6^3D_2$	
18 128	18 130	I	$6p'^3F_4-6^3D_3$	1

[a] In the case of Hg II, levels of the form $5d^{10}(^1S)nx$ are denoted by nx. Other levels are denoted by the "author" notation of Ref. 4.

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[1] J. D. Rigden and A. D. White, Nature **198**, 774 (1963).

[2] R. A. Paananen, C. L. Tang, F. A. Horrigan, and H. Statz, J. Appl. Phys. **34**, 3148 (1963).

[3] W. E. Bell, Appl. Phys. Letters **4**, 34 (1964).

[4] C. E. Moore, *Atomic Energy Levels* (National Bureau of Standards, Washington, 1958), Vol. III, p. 196.

[5] H. M. Crosswhite and G. H. Dieke, *American Institute of Physics Handbook* (McGraw-Hill Book Company, Inc., New York, 1963), 2nd ed., pp. 7-124.

apparent. In the first place, a large number of upper laser levels are clustered around 120 000 cm^{-1} above the ionic ground state; in fact, every doublet in the range 108 000–128 000 cm^{-1} is involved in a transition (Fig. 1). Secondly, in each alkali-like multiplet, only one transition permits laser operation (usually the line of highest multiplicity). Finally, the opposite situation holds in Hg I, where laser action seems to occur in "families" comprising members of the same multiplet.

We believe these results indicate that the primary mechanism populating the upper states in Hg II is direct electron collision from the ground state of Hg. The situation in the lasing levels of Hg I is less clear; it could involve cascades from above as well as direct excitation. In the case of Hg II, excitation may involve large angular momentum transfers, but the transfer of population is predominantly to the $J=L+S$ state. It is true that the line which thus permits laser operation is the one of highest oscillator strength, but the gain of some of these transitions is so high that, if population differences were equal, the next strongest line in each multiplet should also certainly permit laser operation. Nevertheless, the weaker lines are not observed, and we can only conclude that $J=L+S$ states receive a greater influx of population than would be expected from statistical considerations alone. (This does not explain the situation in regard to transitions starting from *s* states, but both of the observed lines are already quite weak.)

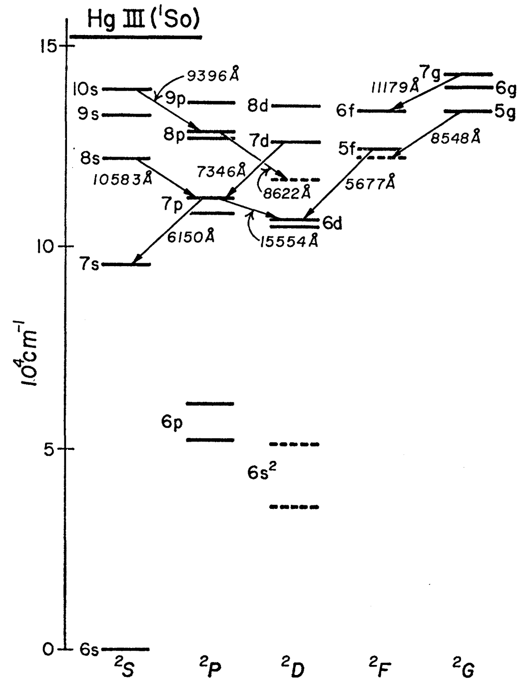

FIG. 1. Partial energy level diagram of Hg II. Levels with excited *d* electrons, when included, are shown with dashed lines. The transitions shown are the observed laser transitions definitely attributable to Hg II.

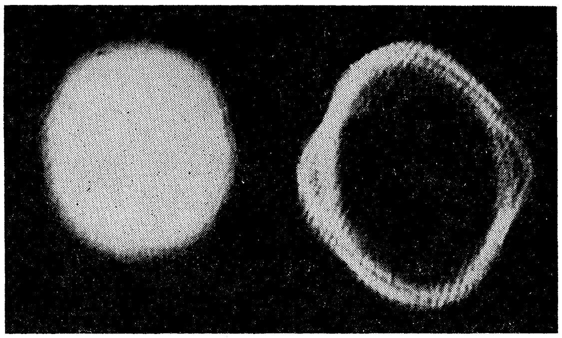

FIG. 2. Laser outputs at 5677 Å (left) and 6150 Å (right). The two wavelengths have been separated by reflection off a grating to permit black-and-white photography.

The green line has also been observed in a mercury-neon discharge. This appears to rule out resonant transfer of energy from metastable atoms as a populating agent, at least in the case of this line.

Further information about the excitation process comes from gain variations in the green (5677 Å) and red (6150 Å) lines as a function of excitation energy. With our present apparatus, we can make minor changes in the electron energy distribution by variations in pulse rise time and current. When the energy is low, only the red line is seen, and when the energy is high, only the green, as would be expected from the difference in upper-state energy values (Fig. 1). For intermediate energies, however, there is a region in which the output of a confocal resonator consists of a green center surrounded by a red ring (Fig. 2). This presumably indicates that electrons near the walls are "cooler" than those in the center, and that population is a fairly sensitive function of electron energy. Further evidence that this is a direct upper-state population phenomenon comes from the experimentally determined fact that when the discharge is such that λ6150 permits laser operation in a ring mode, then λ15554 (which starts from the same upper state) also permits laser operation in a ring mode, whereas other lines permit laser operation in the central spot. Since λ7346 and λ10583 permit laser operation in the central spot, the cascade into the 7*p* state is not in itself sufficient to provide the population inversion for λ6150; an additional direct populating agent is needed.

ACKNOWLEDGMENTS

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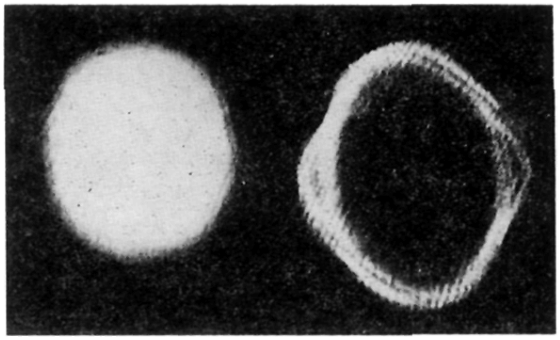

FIG. 2. Laser outputs at 5677 Å (left) and 6150 Å (right). The two wavelengths have been separated by reflection off a grating to permit black-and-white photography.