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Observations of Gain by Stimulated Emission in the Werner Band of Molecular Hydrogen

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Stimulated emission of vacuum ultraviolet radiation has been observed on two $Q1$ lines in the Werner band ($C^1\Pi_u \to X^1\Sigma_g^+$) of molecular hydrogen. These lines, 1161 and 1230 Å, contain 5 kW peak power and were produced by a traveling-wave discharge system. Using this excitation principle, evidence of gain has been obtained by observations of optimum velocity matching at pulse velocities less than c.

Observation and verification of amplification by the stimulated emission of radiation at photon energies above 10 eV have been made for the first time. The availability of such short-wavelength photons in an intense, directional beam is of immediate importance particularly in such fields as photochemistry, photofragment spectroscopy, and photoionization. Stimulated emission of these highly energetic photons has been produced in molecular hydrogen by rapid inversion of the vibrational levels of the $C^1\Pi_u$ excited electronic state with respect to the upper vibrational levels of the $X^1\Sigma_g^+$ ground state.

Speculation on the possibility of laser emission from the Werner band ($C^1\Pi_u$-$X^1\Sigma_g^+$ was first published by Bazhulin, Knyazev, and Petrash.[1] Figure 1 shows a greatly simplified energy-level diagram for the hydrogen molecule. A fast-rising current can produce an electron energy distribution which will preferentially populate the upper electronic states, inverting the vibrational levels with respect to the upper vibrational levels of the $X^1\Sigma_g^+$ ground state. A detailed rate-equation analysis of the Werner band for such a discharge was carried out by Ali and Kolb.[2] This analysis predicted lasing on several vibrational transitions with peak power density of about 20 kW/cm^3 for the strongest vibrational band. It

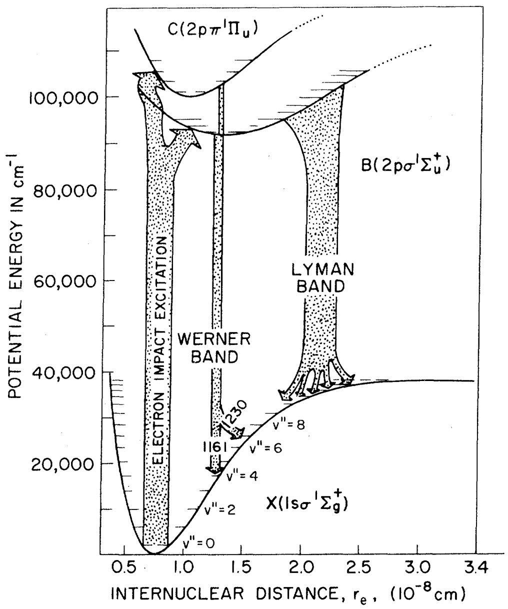

FIG. 1. Energy levels of H_2 from which lasing has been observed.

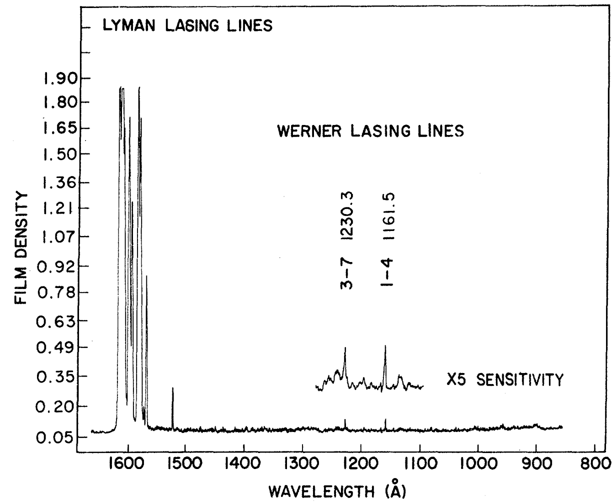

FIG. 2. Densitometer trace showing both Lyman- and Werner-band lasing lines. The new Werner-band lines are shown on an increased sensitivity scale for added clarity; the smaller linelike structure shown on this scale is film noise and not additional emission.

must be realized that this power would be divided among several rotational lines.

Experimentally, vacuum-ultraviolet laser action in H_2 was observed initially in the Lyman band at 1600 Å.[3,4] The traveling-wave discharge system of Shipman had been used previously[3] in the Lyman band to produce total peak powers of several megawatts in a short (~1 nsec) pulse. This traveling-wave system, with several lossy optical components removed from the vacuum optical transmission path, has been used to generate the shorter-wavelength (1161–1230 Å) Werner-band lasing first reported here. These higher-energy photons were detected by passsing the unfocused laser emission through a 1.5-m, 10-mm-diam vacuum path and then through the 0.025-mm slits of a 1-m vacuum monochromator equipped with a film holder and a 600-line/mm platinum-coated grating. Kodak 101-01 film was used to record the superimposed emission from several pulses, and a microdensitometer trace for the 800–1600-Å region is presented in Fig. 2. The monochromator was wavelength calibrated in the first order using argon and atomic hydrogen lines. The identified lines have no more than a ±0.3-Å uncertainty when examined by comparator.

The traveling-wave excitation system explained in detail previously[3] has the capability of varying the propagation velocity of the excitation wave longitudinally down the laser channel. Casperson and Yariv[5] have recently shown that the laser-pulse propagation velocity is gain dependent and is less than c when gain is present. It was previously shown[6] experimentally that maximum laser output was obtained both in N_2 and in the Lyman band of H_2 when the excitation wave velocity was less than c. This velocity-matching technique has now been extended to the newly observed Werner-band lines. It has been seen that these lines do not appear on film when the excitation wave travels at a phase velocity of c or greater, but do appear for excitation phase velocities less than c. This output enhancement by velocity matching is taken as evidence that the pulse traveling in the excited (inverted) gas region is being amplified, is traveling at a velocity less than c, and is showing a greater intensity for $v_p < c$ because of a reduction in spontaneous-emission losses which occur between the excitation wavefront and the laser pulse.

Estimates of the peak power in the two Werner lines were made by comparing the film densities with those of the Lyman band, where power has been measured more accurately by a photoelectric technique. These lines each appear to contain about 5 kW, based on the assumption that the pulse width remains about 1 nsec at these shorter wavelengths. Direct pulse-width measurements have been hampered by the low photon flux available after wavelength selection and by the lack of fast, sensitive detectors in this wavelength region. The 5-kW peak power observed here was emitted from a 36-cm^3 volume of H_2 at 20 Torr pressure. The measurement was made through a LiF window which can be expected to attenuate 50% or more at these wavelengths. This corresponds to an observed power density of 250–300 W cm^{-3} or more.

The identification of the two new Werner-band lines was facilitated by using the Franck-Condon factors calculated by Spindler.[7] If the Franck-Condon factor for exciting transitions from the $v''=0$ vibrational level of the $X^1\Sigma_g^+$ state to any vibrational level of the $C^1\Pi_u$ state (i.e., $q_{0v'}$) is multiplied by the Franck-Condon factor for the emissive transition from v' of $C^1\Pi_u$ to v'' of $X^1\Sigma_g^+$ (i.e., $q_{v'v''}$), the product $q_{0v'}q_{v'v''}$ gives a relative strength of the emission within the Werner band. Table I shows the vibrational transitions and the product $q_{0v'}q_{v'v''}$ in numerical order of the expected intensity for the nine most likely laser levels. As shown, the 1-4 transition would be expected to be the strongest followed by 2-5, 3-7, and 2-6.

Rotational levels must also be taken into account. Since 66% of the molecules are in the $J''=1$ rotational level at room temperature, the upper rotational levels $J'=1$ and $J'=2$ would be populated most heavily by excitation. For transitions involving $\Delta\Lambda=1$, P, Q, and R rotational lines are allowed with Q lines most probable, based on Hönl-London formulas. Table I gives wavelengths for the $Q1$ transitions according to the values given by Richardson.[8] With the use of wavelengths, the two lines observed correspond to the $Q1$ lines of 1-4 and 3-7. The observation of the $Q1$ transitions corresponds to the rotational line expected. Although the $J'=1$ and $J'=2$ levels would receive equal excitation, the $Q1$ ($J'=1 \rightarrow J''=1$) transition dominates during emission according to the Hönl-London formulas. The $R1$, $P3$, and $Q2$ rotational lines all have a lower transition probability and would require a higher gain in order to be observed. The 1-4 transition is observed to be strongest, in agreement with the calculated Franck-Condon factors. The 3-7 transition should, however, appear somewhat less

TABLE I. Relative probability of emission in the Werner band according to the Franck-Condon factors calculated by Spindler (Ref. 7), and the resulting wavelengths of the $Q1$ transition as given by Richardson (Ref. 8).

$v'-v''$	$q_{0v'}q_{v'v''}$	$Q1$ (Å)
1-4	0.0571	1161.32
2-5	0.0546	1175.87
3-7	0.0462	1229.98
2-6	0.0436	1218.94
0-2	0.0414	1099.45
1-3	0.0413	1116.34
0-1	0.0406	1054.18
4-8	0.0380	1239.53
3-6	0.0297	1189.38

intense than 2-5 according to Franck-Condon factors, but it was seen in the Lyman-band laser emission spectrum[3] that the intensities did not strictly obey these guidelines. With increased laser power or detection sensitivity, more of the other lines in Table I are expected to show up.

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