Vacuum-uv Laser Action Observed in H_2 Werner Bands: 1161–1240 Å

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Stimulated emission has been observed in the $Q1$ and some $P3$ lines of the $v' \rightarrow v'' = 1$-4, 2-5, 2-6, and 3-7 Werner bands ($C^1\Pi_u \rightarrow X^1\Sigma_g^+$) of molecular hydrogen near 1200 Å. Light pulses of approximately 1 nsec duration were produced by bombarding hydrogen gas with ~400-keV electrons from a commercial electron-beam generator.

Observations of laser action in the vacuum-ultraviolet spectral region near 1600 Å have been published.[1-3] This Letter reports stimulated emission of even more energetic transitions in the H_2 Werner bands ($C^1\Pi_u$-$X^1\Sigma_g^+$). The wavelengths of the light, from 1161 to 1240 Å, are the shortest laser wavelengths achieved to this time, and the ~10-eV photons can be used for photochemistry, photodissociation, and photoionization investigations of most molecules.

The threshold power needed for lasing action in the vacuum-uv increases drastically as the frequency is raised. In order to introduce high power into hydrogen gas on a nonosecond time scale, the devices used[1,2] to produce lasing near 1600 Å were essentially capacitors, strip lines, and discharge tubes optimized for low inductance. Unfortunately, these devices did not excite sufficient molecules to the $C^1\Pi_u$ state to show high gain and super-radiant laser action on the Werner-band system. In addition, the device described in Ref. 1 has not been engineered to be a reliable laboratory instrument. For these reasons, a field-emission electron-beam generator[4] which delivers 5×10^9 W in a 3-nsec pulse is presently used as a power source for shorter-wavelength lasers.

Figure 1 shows a sketch of the apparatus which produces stimulated emission on the H_2 Werner bands. A 1-cm-i.d. stainless steel tube is bolted to the front face of a Febetron[4] 706 electron-beam generator. The generator consists of a Marx circuit followed by a Blumlein-circuit voltage doubler and pulse sharpener which delivers a 600-kV-peak pulse to a 50-Ω load. The voltage is applied between a number of etched tungsten needles and a 1-mil-thick Ti anode. Electrons are pulled off the cathode by field emission and accelerated across the vacuum gap to the anode. They pass through the anode (losing about 10% of their energy) and are available to excite and ionize the hydrogen gas at a pressure of 20–100 Torr in the tube.

A pulsed magnetic field of 6 kG confines the electrons to a small cross-sectional area in the line of sight. An electron-beam current of ~ 4000 A/cm^2 follows the field lines to the end of the field coil 2.3 m away from the anode, where it diverges into the tube wall 20 cm away from the entrance slit of the vacuum monochromator.

A slightly different version of this setup has already been successful in producing laser action in the H_2 Lyman bands,[5,6] the N_2 laser system at 3371 Å,[7] and in a number of lines of Ne I and Ne II.[8] It may also be commented that the present improvements over the system described in Ref. 6 increased laser powers to ~400 kW/cm^2 on several of the Lyman-band lines.

In all these electron-beam–pumped gas lasers, the high-energy primary, secondary, and cascade electrons induce transitions between the ground

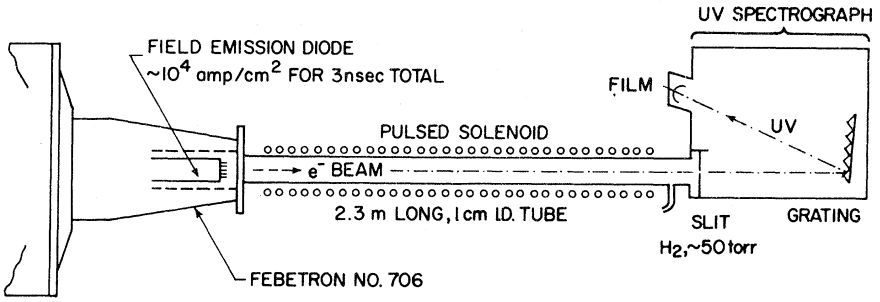

FIG. 1. Sketch of experimental arrangement. The 1-m McPherson monochromator is equipped with a 1200-line/mm Al + MgF_2 coated grating blazed at 1500 Å. Spectra are recorded using Kodak Pathé SC 7 film.

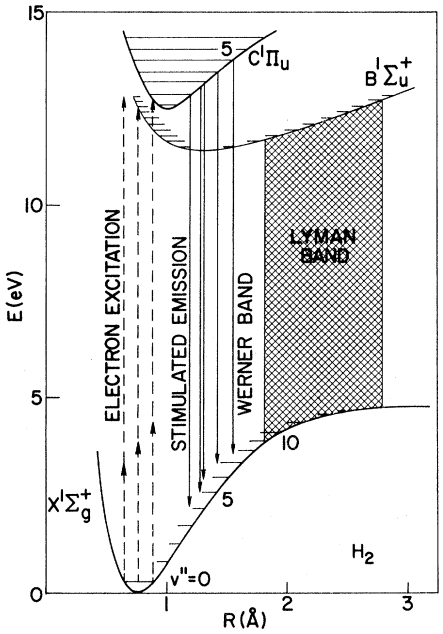

FIG. 2. Potential-energy curves for the $X^1\Sigma_g^+$, $B^1\Sigma_u^+$, and $C^1\Pi_u$ states of H_2. The dashed vertical lines represent the electron-collision–induced transitions between the zeroth vibrational level of the ground molecular state and the v' levels of the upper states. Solid vertical lines indicate the stimulated transitions of the Werner bands. The cross-hatched area is the region of previously observed stimulated emission in the Lyman bands; see Refs. 1 and 2.

electronic state and the excited electronic states. In H_2 the (v') vibrational levels of the $B^1\Sigma_u^+$ and $C^1\Pi_u$ states are excited more efficiently than the upper vibrational levels of the ground electronic state. These upward transitions are indicated by the dashed vertical lines on the potential-energy diagram (Fig. 2). The very high-power, nanosecond electron pulse produces a large inversion density before the spontaneous emission can populate the upper vibrational levels of the ground electronic state. The gain is then high enough for stimulated emission (without mirrors) on the downward transitions indicated by the solid lines of Fig. 2.

The principal diagnostic method used for the present work is spectral analysis of the light emitted from the H_2 under electron-beam bombardment. Figure 3 shows a microdensitometer trace of the Werner-band emission spectrum with sufficient resolution (0.25 Å) to resolve most of the lines of the emission spectrum. The relative wavelengths of the lines agree to within the 0.05-Å measuring error with Jeppeson's[9] measured wavelength for the assignments. The

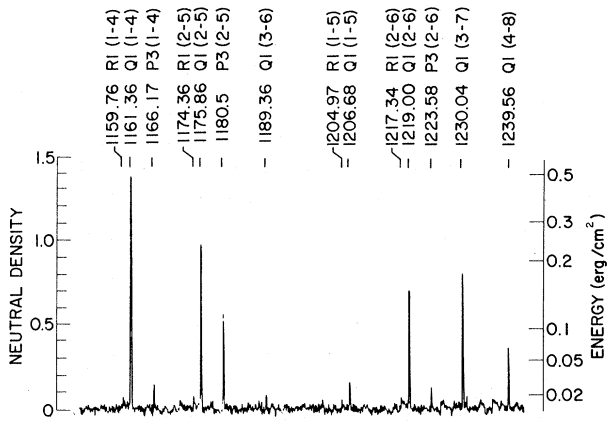

FIG. 3. Microdensitometer trace of the Werner-band stimulated-emission spectrum. The measured neutral density is plotted on the left as a function of the wavelength, and the corresponding film exposure in erg/cm^2 calculated from Ref. 10 is indicated on the right.

spectrum shown in Fig. 3 was exposed in a single shot with no window between the tube and the film so that the spectrum below 1050 Å (the cutoff of the LiF window) could be exposed. Spectra exposed with the window in place are identical to that in Fig. 3, and are completely different from the normal spontaneous emission spectra. It will be shown later that this difference proves that stimulated emission, and not spontaneous emission, is occurring.

From the neutral densities of the lines recorded on the left side of Fig. 3, the exposures (in erg/cm^2) are calculated using the work of Burton, Hatter, and Ridgeley.[10] Assuming the same 1-nsec pulse length measured[7] for N_2 or H_2 (near 1600 Å), these measured exposures indicate power densities of up to 50 W/cm^2. In fact, subsequent exposures gave maximum powers of 500 W/cm^2 in the (1-4) $Q1$ line. These estimates are quite conservative, since the efficiency of the 1200-line/mm grating and the LiF-window losses have not been taken into account.

The power output from an electron-beam–pumped gas laser can be estimated by estimating the inversion number density and calculating the gain. This was done by assuming that the ratio of excited-electronic-state density to ion density equals the ratio of excitation to ionization collision cross sections[11,12] for high-energy electrons, and by using measured ion production coefficients.[13] An inversion number density of 6×10^{11} was obtained this way, which indicates a relatively low gain of 7×10^{-2} cm^{-1} for the strongest transition. We can calculate that the energy should be about

10^3 erg/cm^2 with this gain. This is significantly greater than the maximum observed energy of 5 erg/cm^2. However, with the present near threshold situation, the calculated energy depends exponentially upon the estimated gain, and the disagreement is not too surprising. The difference does call attention to the fact that significant increases in the present output are theoretically feasible.

For comparison with the above laser energy, the energy density from spontaneous emission falling on the spectrograph slit is calculated to be <0.05 erg/cm^2 for the (1-4) $Q1$ line. Exposure of some of the weakest lines shown in Fig. 3 may well be due to spontaneous emission.

The stimulated-emission power available is not higher because most electrons lose only a negligible fraction of their energy before they hit the tube walls. As the pressure is raised to increase this fraction, evidence of beam instability is noted[7] and the laser power drops. If this problem can be overcome, the new developments in electron-beam technology can be applied to make these electron-beam pumped gas lasers relatively convenient and useful.

Stimulated emission of several transitions of the Werner-band system can be proven by a variation of a "spectral-line-narrowing" argument. The two energy levels of the $C^1\Pi_u$ state expected on the basis of intensity rules[14] to be most highly populated are the $J'=1$ lower Λ level and the $J'=2$ upper Λ level of the $v'=1$ vibrational state (Fig. 4). The branching ratios for spontaneous emission from these states are determined[14] by the v'-v'' Franck-Condon factors,[15] the selection rules, and the statistical weights of the rotational levels. Only transitions with $\Delta J=0$ (Q branch lines) are allowed from the lower Λ levels, while only transitions with $\Delta J=+1$ and -1 (P and R branch lines) are allowed from the upper Λ levels.[14]

The $Q1$ line of the $v'=1$ to $v''=4$ band is expected[14,15] to be 1.4 times as intense as another transition from the same upper state, the $Q1$ line of the 1-3 band at 1116 Å. However, in the spectrum recorded, the 1-4-band $Q1$ line is at least 50 times as intense as the 1-3-band line, which is too weak to detect. The only explanation for such anomalous intensity ratios is that the $v'=1$, $J'=1$ lower Λ level has been stimulated to emit on the higher gain (1-4) $Q1$ transition.

A different example, again on the 1-4 band, can be seen by referring to Fig. 4. The $Q1$ transition gain is expected[14] to be $\frac{5}{3}$ as great as the gain on the $R1$ transition, and $\frac{5}{2}$ as great as the gain on the $P3$ transition from the $J'=2$ upper Λ level. If, in fact, the $Q1$ emission is stimulated, the $J''=1$ level will "fill up" and the population inversion and gain on the $R1$ transition will be reduced. The $J'=2$ upper Λ level will then at some point be stimulated to emit on the $P3$ transition. Figure 3 shows, for example, that the $P3$ line of the 1-4 band is at least 3 times more intense than the $R1$ line. In spontaneous emission, the $R1$ line is the stronger in the ratio 3:2. Such an altered intensity ratio is even more striking in the 2-5 band where the $P3$:$R1$-line intensity ratio is greater than 10:1.

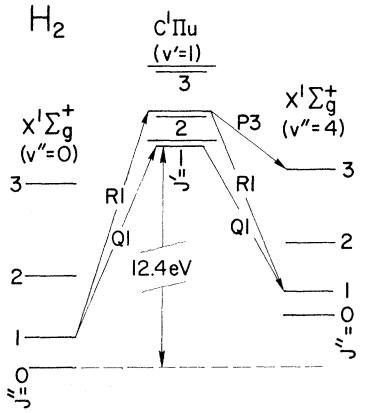

FIG. 4. Schematic energy-level diagram for the rotational sublevels of the $X^1\Sigma_g^+$ ($v''=0,4$) and $C^1\Pi_u$ ($v'=1$) vibrational levels. For clarity, the Λ-type doubling of the $^1\Pi_u$ state has been greatly exaggerated and the energy scale of each vibrational level has been shifted.

No trace is seen of the $P3$ line of the strong 3-7 band. The $P3$ and $R1$ lines are known to be perturbed by a level of the $B^1\Sigma_u^+$ state, and the effect seems to be identical to the stimulated emission of the CO fourth positive bands,[16] where sets of perturbed rotational lines are missing.

In summary, we have shown that stimulated emission of the shortest-wavelength (1161 Å) laser light reported to date can be produced using an electron beam to pump H_2 gas.

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[1]R. T. Hodgson, Phys. Rev. Lett. 25, 494 (1970).
[2]R. W. Waynant, J. D. Shipman, Jr., R. C. Elton, and A. W. Ali, Appl. Phys. Lett. 17, 383 (1970).
[3]N. G. Basov, V. A. Danilychev, Yu. M. Popov, and D. D. Khodkevich, Pis'ma Zh. Eksp. Teor. Fiz. 12,

473 (1970) [JETP Lett. 12, 329 (1970)].
[4]Field Emission Corp., McMinnville, Ore.
[5]R. T. Hodgson and R. W. Dreyfus, in Proceedings of the Twenty-Fourth Gaseous Electronics Conference, Gainesville, Florida, 5-8 October 1971 (to be published).
[6]R. T. Hodgson and R. W. Dreyfus, "Electron-Beam Excitation of a Vacuum-Ultraviolet Hydrogen Laser" (to be published).
[7]R. W. Dreyfus and R. T. Hodgson, Appl. Phys. Lett. 20, 195 (1972); M. Clerc and M. Schmidt, C. R. Acad. Sci., Ser. B 272, 668 (1971).
[8]R. W. Dreyfus and R. T. Hodgson, to be published.
[9]C. R. Jeppeson, Phys. Rev. 44, 165 (1933).
[10]W. M. Burton, A. T. Hatter, and A. Ridgeley, Amer. Astronaut. Soc. Photo-Bull. 1, 27 (1971).
[11]E. J. Stone and E. C. Zipf, to be published.
[12]H. S. W. Massey, *Electronic and Ionic Impact Phenomena* (Oxford Univ. Press, Oxford, England, 1969), Vol. II, p. 910.
[13]R. D. Evans, *The Atomic Nucleus* (McGraw-Hill, New York, 1955), Chap. 18, Eq. 2.22.
[14]G. Herzberg, *Molecular Spectra and Molecular Structure: I, Spectra of Diatomic Molecules* (Van Nostrand, Princeton, N. J., 1950).
[15]R. J. Spindler, Jr., J. Quant. Spectrosc. Radiat. Transfer 9, 627 (1969).
[16]R. T. Hodgson, J. Chem. Phys. 55, 5378 (1971).

Theory of the Liquid-Solid Phase Transition in Helium II†

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The liquid-solid phase transition of helium at low temperatures is shown to result from the instability of the roton mode. In a simple model of the roton-roton interactions, the instability arises at a *finite* value of the roton energy as a result of the hybridization of a single roton with a bound roton pair. Thus our model exhibits a discontinuous jump of the soft-mode energy at the phase transition in agreement with neutron scattering experiments.

The stability of a system with respect to density fluctuations is related to the analytic properties of the density-density correlation function $S(k, \omega)$ in a well-known way.[1] At very low temperatures the correlation function is $S(k, \omega) = -\chi''(k, \omega)/\pi$, where χ'' is the dissipative part of the density-density response function $\chi(k, \omega)$. Stability requires that $S(k, \omega)$ be positive for all real frequencies. In other words, poles or other singularities of $\chi(k, \omega)$ cannot appear in the upper half-plane of the ω variable for a stable system, because they signify that the response of the system to an external perturbation of the appropriate frequency ω and momentum k will grow without limit.

The location of the poles in $\chi(k, \omega)$ gives the frequency and damping of the excitation modes in the system. Phase transitions occur when one of these modes becomes soft, i.e., the mode frequency decreases as the transition is approached. In the case of second-order phase transitions, the frequency of the soft mode vanishes at the transition. However, in many first-order phase transitions, such as the liquid-solid transition, it is observed that a mode softens, but the transition occurs well before the mode frequency vanishes.

Recently, it has been suggested[2] that the superfluid-solid phase transition in helium is associated with the softening of the roton mode. In Ref. 2 it is conjectured that the roton energy should decrease with increasing pressure and vanish at the stability limit. The roton is a natural candidate for the soft mode in liquid helium since its wave vector k_0 is close to the reciprocal-lattice vector of solid helium.[2]

Recent neutron scattering experiments[3,4] on liquid helium at various pressures show that the roton energy decreases with increasing pressure, but does not tend to vanish as the solid phase is approached! At $T \simeq 1$°K the minimum roton energy Δ is observed to decrease smoothly from $\Delta = 8.65$°K at saturated vapor pressure to $\Delta = 7.0$°K at $P = 25.3$ atm.[3,4] Since the latter pressure is very close to the pressure needed to solidify the helium, it is apparent that the roton energy does not approach zero at the slightly higher phase-boundary pressure. To our knowledge the physical origin of the roton softening and instability under pressure has not been explained from the theoretical point of view.

In the present paper we present a calculation