Excitation mechanisms of the e-beam–excited hydrogen laser*

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We have studied the e-beam–excited molecular-hydrogen laser and the effects of foreign gases, preionization, and cooling the gas. The pressure dependence up to 200 Torr of the laser output and the influence of the impurity gas are described in terms of the energy distribution of the plasma electrons produced in the gas breakdown due to the electric field induced by the fast-rising current pulse. Small additions of argon and nitrogen sharply reduce the laser output, unlike helium which has little effect. Preionization of the gas destroys the lasing action which fully recovers in a time of the order of a millisecond. High-repetition-rate three-level laser action should be possible. Magnetic neutralization of the induced electric field or destruction of inversion would both account for the loss of laser action. The optical gain is increased to more than 300% by liquid-nitrogen cooling. The gain depends on the reduction of Doppler broadening by cooling but must also be understood in terms of the thermal redistribution of the rotational populations. Arguments are given in favor of the direct electron excitation of lasing levels in contrast to excitation involving charged intermediate states.

I. INTRODUCTION

In recent years there has been considerable interest in ultraviolet and vacuum-ultraviolet (VUV) lasers.[1] Following the field we refer to the amplified spontaneous emission as laser behavior when it is observed in molecular hydrogen. This lasing action in the VUV spectral region of hydrogen has been produced by transverse discharge[2–4] and by relativistic electron beams.[5–8]

Several excitation models have been proposed to describe the inversion process. Dreyfus and Hodgson[7,8] have suggested that in e-beam experiments the excitation of hydrogen molecules for lasing action is produced by primary, secondary, and cascade electrons. On the other hand, for the e-beam–excited nitrogen laser, McArthur and Poukey[9] have demonstrated that low-energy plasma electrons are responsible for most of the molecular excitation. In the transverse-discharge experiments the plasma electron model is ordinarily assumed with considerable success. Although direct excitation models are most widespread, Gallardo, Massone, and Garavaglia[10] suggested that the temporary negative ion H_2^- formed by excitation in the 11–14-eV region would decay into the upper lasing states of molecular hydrogen and that this process would lead to the inversion. Recent studies of lasing action in high-pressure hydrogen[11,12] are not directly relevant to the present work since, at high pressure, four-level laser action and collisional predissociation provide the framework for discussion.

All excitation models depend strongly on the energy distribution of the electrons within the gas. In turn, that distribution will depend on the production mechanism for the electrons. A fundamental difference between the mechanisms of excitation by plasma electrons and by secondary or cascade electrons involves the electron energy distribution in the presence of large electric fields or without invoking such fields, respectively.

It was pointed out by McArthur and Poukey[9] that the plasma current and the characteristic energy of electrons vary considerably as the drift-tube radius changes and therefore a check of the importance of plasma-electron excitation would be the laser-output measurement as the radius of the gas cell was changed. Putnam's calculation[13] also shows that the induced electric field depends on the ratio of the tube radius to the beam radius owing to the boundary conditions in the conducting walls of the gas cell. Evidence on this point for the hydrogen laser may be found in the reports of Hodgson and Dreyfus.[5,8] The optimum pressure for gas cells of two differing radii was found to be significantly smaller for the smaller of the two gas cells when the experimental conditions apparently were otherwise quite similar. This trend is in the direction expected for the plasma-electron excitation model but is difficult to understand from the cascade-electron model. Since the authors were not deliberately checking the dependence of output on radius, this evidence is only suggestive.

The experiments reported here are intended to give further evidence on the mechanism of excitation. We have investigated the influence of some foreign gases and of preionization of the hydrogen prior to e-beam injection. The primary measurements here are the determination of the relative laser output as a function of gas pressure

and temperature, or of the time delay between the preionization and e-beam pulses.

The success of the McArthur-Poukey model for the e-beam–excited nitrogen laser makes it plausible to assume the same model for hydrogen and to perform the suggestive tests described here. The following sections include a brief description of the apparatus and techniques followed by results and discussion.

II. APPARATUS

The machine used in our experiment was a model 105 Pulserad field-emission accelerator.[14] The output in the electron beam is normally 700 keV, 10 kA, with a pulsewidth of about 20 nsec and a risetime of about 10 nsec. The electron-beam diameter in the gas cell was somewhat less than 1 cm when a magnetic guide field was used. The accelerator consists of a Marx generator and a coaxial pulse-forming Blumlein transmission line. The high voltage was applied between a hollow-tipped metal cathode and a 25-μm-thick aluminum foil in the optimum configuration. The foil is grounded and separates the evacuated diode section and the gas cell. The foil was destroyed each time the accelerator was fired. Many cathode shapes, foil thicknesses, and material variations were studied. The gas cell is similar to that of Hodgson and Dreyfus[5] and is made of a stainless-steel tube about $3\frac{1}{2}$ cm in inner diameter and about 104 cm long. A solenoid wound directly on the tube provides the pulsed, external magnetic field up to 20 kG, energized by a 96 μF 10-kV capacitor bank, to guide the field-emitted electron beam down the gas cell. Maximum laser output was found with a 9-kG field. The e-beam current could be monitored with a Rogowski coil placed directly behind the anode foil. Research-grade hydrogen produced by Matheson Gas Products, Inc. was used throughout the experiment, but lower-purity grades gave the same results. Details of synchronization and variations of output with magnetic field are described elsewhere.[15] Cooling was facilitated by a styrofoam box fitted around the gas cell which would then be totally immersed in liquid nitrogen.

For preionization we used a glass tube with high voltage and trigger electrodes inserted transversely into the tube as shown schematically in Fig. 1. The high-voltage feedthrough electrode, located approximately 35 cm from the metal cylinder, initiated a discharge toward the metal cylinder. Because the pulsed external magnetic field always triggered the discharge by itself when high voltage was applied to the hydrogen, we did not use the magnetic field for this part of the experiment. Although comparable energies were deposited in the gas by preionization and by the e beam, as expected the former never led to laser action by itself. A capacitor bank of 4 μF was used to predischarge the gas; the charging voltage was 7–10 kV. The gas pressure was 20–30 Torr. The discharge occurred between the pointed feedthrough electrode and the sharp edge of a metal cylinder inserted inside the glass tube behind the foil holder.

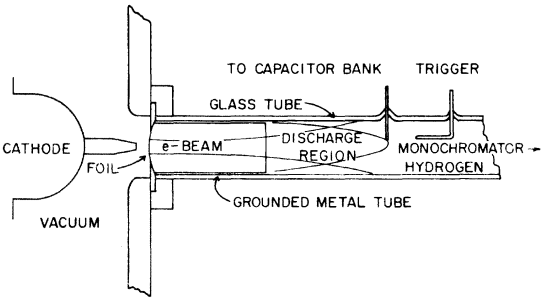

FIG. 1. Schematic view of the gas cell used in preionization experiments. In the absence of a guide field the e beam was confined to the discharge region whether preionization occurred or not.

When the preionization discharge took place, only the region near the e-beam inlet was preionized. In addition, in the absence of a guide field, the e beam propagated only a short distance beyond the metal cylinder which was inserted into the glass tube. Thus the regions of preionization and beam propagation roughly coincided.

We believe that lasing action took place only in the volume enclosed by the metal cylinder when the gas was preionized. A Rogowski coil around the glass tube between the electrode and the metal insert permitted the monitoring of the e-beam current as well as of the currents arising from the preionization. No evidence of important change in the propagation was found, nor correlation with the time delay between preionization and e-beam injection. In the absence of the guide field the volume of hydrogen excited was marginally successful for lasing action owing to the poor propagation of the e beam. This propagation was so poor that the preionization apparently did not represent a serious perturbation although changes in propagation would importantly change the laser output.

A high-vacuum, 1-m Seya-Namioka monochromator equipped with a grating of 600 lines/mm, blazed at about 1500 Å, with aluminum-MgF_2 overcoating, was used to disperse the laser spectrum which was photographed. A LiF or MgF_2 window

(1 mm thick) positioned behind the 25-μm entrance slit usually separated the gas cell from the evacuated monochromator. Calibration tests without a window were easily made by allowing hydrogen to fill the monochromator. This procedure risked damage to the grating by high-speed aluminum bits from the target foil so the window was customarily inserted. The exit arm of the monochromator had a wavelength shifter (vacuum ultraviolet to visible) consisting of a glass window coated with sodium salicylate to a density of about 1 mg/cm^2. For photographing the spectrum we used Kodak Royal-Xpan films 4166 (ESTAR thick base, 4×5 in.), which have a speed ASA 1250. By increasing the recommended development times by about 50%, ASA 4000 can be obtained. We made no attempt to measure the absolute flux of light. Careful measurements in a similar system were made by Dreyfus and Hodgson.[8] Our pulses were surely less intense (~10^{12} photons/cm^2) owing to our short gas cell, whose length was limited by the length of the lead-shielded room available for the experiments. The exposed spectrograph was scanned with a calibrated Joyce Loebl & Co. automatic recording microdensitometer. Separate experiments were used to establish the relative-intensity–optical-density calibrations for our development procedure.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The spectra of the Lyman-band lasers observed in our experiment are very similar to those reported by Hodgson,[2] Waynant *et al.*,[16] and Antonov *et al.*[11] although the excitation techniques are quite different. These groups used transverse discharge in the gas. The spectra are very similar in atmospheric-pressure hydrogen[11,12] excited transversely and in e-beam excitation of low-pressure hydrogen.[5,8] These facts do not imply that the same excitation mechanism is involved since the character of the spectrum is dominated by Franck-Condon (FC) factors.

First we will consider the relative intensities of some of the lines of the Lyman-band laser. Only some P branches of a few v'-v'' progressions consistent with the FC principle appear in the spectrum, namely $P(1)$, $P(2)$, and $P(3)$. This can be understood from the fact that most of the hydrogen molecules at room temperature are in $J''=0$ (14%) and $J''=1$ (66%) of the ground state, $^1\Sigma_g^+$ ($v''=0$), and that the dipole-transition selection rule, $\Delta J=\pm 1$, should hold in direct electron excitation. The $P(1)$ and $P(3)$ branches originate from the $J''=1$ level and the $P(2)$ from the $J''=0$ level of the ground state. The $R(1)$ branch, although never seen to lase, is also possible, but it is probably suppressed in lasing since it originates from the same level as $P(3)$. One interesting point, as roughly understood from their common X state origin, is that in each progression the $P(3)$ line is stronger than the $P(1)$ line as predicted by Ali and Kepple.[17] The $P(2)$ line is least intense whenever it lases, probably because the initial population of the $J''=0$ level of the ground state is only about one-fifth of that of the $J''=1$ level. The retention of information of ground-state rotational populations through excited-B-state levels back to the final state of the lasing transition supports the direct-excitation mechanism. Below we will also argue further that the temperature dependence within these progressions suggests a direct-excitation mechanism. Owing to the low intensity in the Werner band produced in our short gas cell, we were only able to observe the Werner-band laser at liquid-nitrogen temperature.

The optimum pressure for the laser output in molecular hydrogen has been reported by Waynant *et al.*[16] and Hodgson and Dreyfus[2,5,8] in the 20–50-Torr range. We show in Fig. 2 the pressure dependence of the laser line $P(3)$ (5-12) 1613 Å in the Lyman band at room temperature in our experiment. We observed that the lasing action takes place from about 10 Torr to more than 200 Torr, peaking at around 50 Torr. We did not see lasing action at pressures higher than 200 Torr up to 1500 Torr. The general character of this pressure dependence is very similar to that calculated for N_2 by McArthur and Poukey. Shot-to-

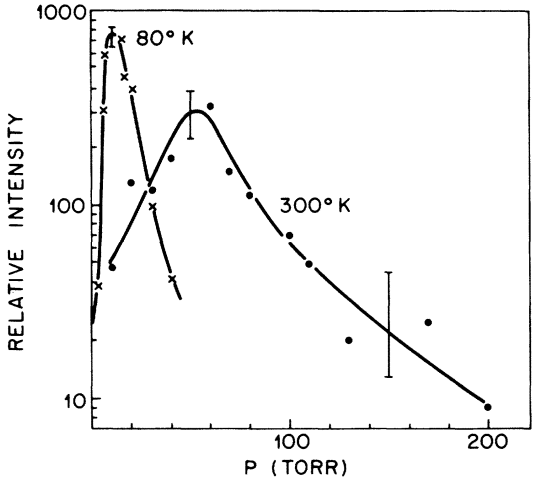

FIG. 2. Relative intensity of the $P(3)$ (5-12) 1613.2-Å Lyman line vs. logarithm of pressure at 300 K (circles) and 80 K (crosses), respectively. The external guide field is 8–9 kG. The solid curves were drawn to guide the eye and have no theoretical basis. Typical error bars based on shot-to-shot variation are indicated.

shot variability is suggested by the error scale indicated in the figure. At pressures greater than 200 Torr in e-beam systems the failure to produce three-level laser action is to be understood from the pumping mechanism. Here we will consider low-pressure behavior where a small portion of the primary beam energy is lost in the region of lasing action.

It is generally accepted[18] that in the transverse discharge the plasma electrons are directly responsible for exciting the gas molecules by electron-exchange collisions. In principle, one can analyze the system to find the α/N value corresponding to a given E/N (α is the Townsend ionization coefficient of the gas, E is the external electric field, and N is the gas number density) and the drift velocity of the plasma electrons, making it possible to calculate the electron density and temperature. Then using the experimental cross sections the excitation rates may be calculated for the laser levels.[18] An optimum pressure for three-level laser output has a clear origin in the plasma-excitation models.[9] At lower pressure than optimum, E/N is larger and ionization is relatively enhanced at the resulting higher electron temperature at the expense of excitation. At higher pressure than optimum, energy is preferentially dumped into the vibrational system.

If cascade and secondary electrons are primarily responsible for excitation, it is difficult to understand why the optimum pressure should be at such a low pressure as 25–50 Torr where the loss rate is very low. In the case of the nitrogen laser, calculations of laser output based on the hypothesis of cascade excitation[19] at the (experimental) optimum pressure were not felt to agree well with experiment. The plasma electron model was more successful.[9] Now in the case of hydrogen it appears plausible to examine implications of the same model. The fractional power input to elastic and inelastic collisions for H_2 has been studied as a function of the characteristic energy of the low-energy electrons in the gas[20] and the characteristic energy for H_2 has been measured as a function of E/N.[21] At a different characteristic energy or a different E/N, a different type of excitation is preferred. For example, at the characteristic energy of 1 eV almost all the power goes into the vibrational excitation, and at around 6 eV the power primarily goes into electronic excitation. It is the electrons in the distribution with kinetic energies near 30 eV which are most effective in producing electronic excitation. At higher characteristic electron energies, ionization becomes predominant. It is E/N that controls the rate of excitation of the electronic states through the effects on the electron temperature in the plasma electron model. We will try to estimate the characteristic energy of electrons using the plasma electron model in the present circumstances. Using Putnam's model[13] the induced electric field may be estimated to be about 3.5×10^3 V/cm. For a hydrogen pressure of 50 Torr this implies[20] a characteristic energy of about 5 eV at which the laser intensity peaked in our experiment. This is in good agreement with the theory that the electronic excitation is most efficient at the characteristic energy of around 6 eV.[20] We are unable to find any argument to produce an optimum pressure of near 50 Torr by assuming that cascade electrons are responsible for the excitation of hydrogen. It is appropriate to look at further experimental evidence which relates to the inversion and excitation mechanisms.

First, we examine the pressure dependence of the relative intensity of the laser output of the $P(3)$ (5-12) line as a function of the partial pressure of admixture gases. This line is the most intense Lyman transition owing primarily to Franck-Condon and statistical-weight factors. Figure 3 shows this output as argon, nitrogen, and helium are added to hydrogen which is held at 30 Torr. Of particular interest is the comparison of the influence of helium and that of argon. The light intensity decreases extremely rapidly with the argon pressure, but it is only weakly dependent on the helium pressure. In the present low-pressure range and the time scale

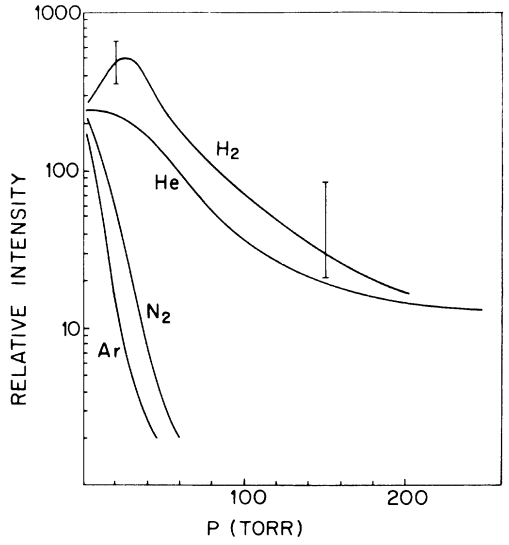

FIG. 3. Relative logarithm of intensity of the $P(3)$ (5-12) 1613.2-Å Lyman line as a function of partial pressure of admixture gases added to hydrogen at 30 Torr. All the experimental data points are dropped from the figure for clarity. Typical error bars, due to shot-to-shot variations, are indicated at high and low intensities.

involved in the lasing action, the collision time for any energy transfer between the hydrogen molecules and the admixture gas molecules would be too long to affect the lasing action. As a result, the major effects of impurity gases are expected to arise from modifications of the electron temperature and competitive loss mechanisms. These competitive losses are expected to be severe since Ar and N_2 also have electronic levels within the same energy range as the upper laser level for hydrogen. The cross sections and further details will be discussed below.

It is useful to consider the electron energy distribution and characteristic temperature for gas mixtures when discussing the influence of foreign gases on the hydrogen excitation. It might be expected that on adding helium to hydrogen at pressure below the optimum, the electron temperature would drop through the optimum range, thus increasing the output. However, this trend is opposed by competitive power loss in the helium itself. Of course, additional plasma electrons arise owing to the added helium and the opposing trends lead to small dependence on the helium pressure. Since at pressures above the optimum, helium is scarcely worse than added hydrogen itself in reducing the electronic excitation of hydrogen the competition is apparently not severe. The low total cross section of helium for 30-eV electrons relative to Ar or N_2, for example,[22,23] supports this interpretation. A second consideration involves the alteration of the distribution of electrons in energy for a particular characteristic temperature. It is found,[24,25] at lower values of E/N, that helium has a disproportionately large number of electrons compared with Ar in the high-energy tail which is effective for electronic excitation. We might expect from this consideration alone, if the same alteration occurred for the higher E/N values of the present regime, that the electronic excitation of hydrogen would drop more quickly for argon in hydrogen than for helium in hydrogen. It is probable, however, that the reason for the rapid drop of laser output for Ar or N_2 in hydrogen is primarily to be sought in the competitive extraction of energy from the plasma electrons due to their higher total cross sections. Of course, if additional hydrogen is added to the original hydrogen the electron temperature will still gradually drop through the optimum value. However, the added hydrogen, unlike the impurity case, not only produces more electrons but supplies more hydrogen molecules in the ground state for laser excitation without competition. Beyond a certain pressure of added hydrogen, the compensation will not keep up with the decrease in the electron temperature and the excitation rate will go down in all cases. The possible crossing of the H_2+He and the H_2+H_2 curves near 200 Torr may be understood from the electron energy distribution curves. Similar arguments can be used to describe the H_2+N_2 results of Fig. 3.

Many of the above arguments also apply to the ultimate electron temperature of the cascade electrons. We therefore cannot presently state that the impurity effects demonstrate the favored position of the plasma electron model, only that it is a convenient framework from description of the observations.

The preionization experiments yield one definite result with an uncertain interpretation. The pulsed discharge from the capacitor bank dumps 100 J into the hydrogen gas comparable to the energy which is deposited at a later time from the electron beam. In these experiments the discharge lasts for a time of 30–40 μsec as measured with a Rogowski coil surrounding the glass tube. Because the discharge was triggered by application of a pulsed magnetic field, we were not able to use the external magnetic field and only several lines of the Lyman band weakly lased at the optimum pressure of 40–50 Torr. Following the pulsed discharge of the hydrogen, we delayed the electron-beam injection by a variable time period looking for the recovery of the lasing action which was destroyed for short delays between the two depositions of energy. When the electron beam was delayed by 0.5 to 1 msec or more, the lasing action recovered. We will examine the several arguments for the destruction of lasing action below. First we will note that because of the small laser gain without an external magnetic field, the recovery indicates the reestablishment of the marginal inversion within 1 msec. This definite result may have considerable relevance to the possibility of an e-beam laser of high repetition rate in hydrogen, although we were not in a position to extend the experiment beyond two depositions of energy.

Considering the large number of reasons why the laser action could be destroyed by preionization, we found it noteworthy that the recovery time was as short as 1 msec. Those reasons include the following and we are not able to suggest which mechanism is most pronounced. We have already pointed out that a perturbed propagation of the beam which could have reduced the excitation below that critical for lasing was not obvious from our measurements with the Rogowski coil. In addition, a recovery time of 1 msec might imply[26] a gas temperature of up to 800 K. Owing to Doppler broadening,[8] manifestations of such a high temperature might have eliminated the laser action which was marginal. In fact, the recovery

of intensity was essentially complete by 1 msec, but the Doppler broadening effects at a temperature lower than 800 K might have been important in killing the laser action for shorter delay times between preionization and e-beam injection. Perhaps more significantly, owing to the high degree of ionization produced by preionization, magnetic neutralization[27] would short out the accelerating electric field in the plasma electron model preventing the production of enough plasma electrons of sufficient energy to create excitation of the hydrogen molecules. After a sufficient period, preionization would no longer be sufficient to inhibit this mode of excitation. Since we do not know the amount of ionization nor the magnitude of the plasma electric field as a function of time, we can say nothing further. In the plasma electron model and in the cascade model as well, the destruction of inversion following preionization due to recombination of ions or electronic recombination leaving the molecule in high vibrational terms of the ground electronic state would likely persist for time scales of 1 msec[26] and easily account for the magnitude of the recovery time. The preionization result does not clearly select between alternative excitation mechanisms but is qualitatively understandable.

The temperature dependence of laser output has been studied for various gas lasers.[28—31] An increase of 300—400% in the laser intensity of the Lyman band was observed by cooling the hydrogen to 80 K in our experiments. Additional laser lines were also observed as reported previously by Dreyfus and Hodgson,[8] and in our work Werner-band lines were observed only on cooling. The pressure dependence, measured at 80 K is also shown in Fig. 2. As previously observed[8] the peak output at the two temperatures occurs at about the same number density, 1.6×10^{18} cm^{-3} and 1.3×10^{18} cm^{-3} at 300 and 80 K, respectively, in our experiments. The increase in laser output power was attributed earlier[8] to the Doppler effect which is related, through the linewidth, to the optical gain in the amplified emission. Although the gain is primarily a function of $1/\Delta\nu$ or $1/\sqrt{T}$, the dependence on the vibrational and rotational temperatures must also be considered as well as the molecular temperature. We point out another important effect of cooling the gas. It was described above that owing to the distribution of the rotational levels at room temperature, only some P branches appeared in the laser spectrum. If the gas temperature is lowered, the gas molecules redistribute themselves within the rotational levels. At 80 K the distribution would be 58% in $J=0$, 41% in $J=1$, and less than 1% in $J=2$ only if thermal equilibrium were reached. Of course, the prohibition of intercombination between symmetric and antisymmetric states does not allow transition from an even-numbered level to an odd-numbered level on our time scales without the use of a catalyst like active charcoal.[32] Thus the distribution of molecules among the rotational levels of the gound state will be roughly 25% in $J=0$ and 75% in $J=1$ at 80 K, compared to 13% in $J=0$ and 66% in $J=1$ at 300 K. Thus there is an increase in the population of both levels which can enhance the inversion of specific lines upon direct excitation and lead to higher laser output upon cooling. It is interesting to note that parahydrogen molecules are all in the even-numbered rotational levels, and $J''=0$ will be most densely populated while all the odd-numbered levels will be empty. Therefore neither the $P(3)$ nor the $P(1)$ lines will appear in the parahydrogen laser while the $P(2)$ branch will be greatly boosted. This is clearly evident in the published results of Dreyfus and Hodgson.[8] The apparent manifestations of the selection rules throughout the laser process directly support the theory that the laser levels are excited by direct electron impact rather than by way of an intermediate resonant state.[10]

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