Laser oscillation and anisotropic gain in the 1 → 0 vibrational band of optically pumped HF gas

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540 dyn sec/cm² as the laser fluence varied from 1.4 to 90.0 J/cm^2 (see Fig. 4). For the case of 63 μ m of Duco cement and a fluence of 90.0 J/cm^2 , the specific impulse was approximately 1500 dyn sec/cm².

The method of stress generation in the above technique differs from the technique used by Anderholm.¹⁰ In that technique a thin metallic film (on the order of a micron) was sandwiched between two transparent materials. The laser vaporized the metallic film and produced an inertially confined high-temperature high-pressure plasma which drove large-amplitude stress waves into the confining materials. A theoretical discussion of the technique is given in Ref. 11. In the case of the thin volatile coatings, the laser flux penetrates the transparent coating and is partially absorbed in the underlying metal target. The absorption of the laser energy rapidly heats the metal surface. Heat is subsequently transferred by conduction and radiation to the coating. When volatile layers are used, the metal target shows little evidence of any thermal damage. In addition, it was found that, as the curing time of the RTV adhesive was increased, the magnitude of the stress decreased. These results imply that the stress is generated by a high-pressure gas produced by partial decomposition of the volatile transparent layer. The expansion of the trapped vapor accelerated the unvaporized portion of the transparent layer to high velocities, thereby enhancing the momentum delivered to the metal target. The decrease in the rate of increase of stress with laser fluence at the higher fluences shown in Fig. 3 is believed to be the result of proportionally more of the laser energy's being consumed in decomposing the complex vapor

molecules and also in vaporizing more material.

In summary, a simple method has been found for augmenting by more than an order of magnitude the stress and specific impulse when lasers are used to irradiate metal targets. The technique can be easily applied to complex geometries, and, when laser systems with gigawatt outputs are used, stress-wave amplitudes on the order of 10 kbar can be produced. This technique has been used to test the dynamic bond strength of electronic components and to load model cylindrical structures.

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Laser oscillation and anisotropic gain in the $1 \rightarrow 0$ vibrational band of optically pumped HF gas*

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P-branch laser oscillations have been observed in the $v = 1 \rightarrow v = 0$ band of HF gas, optically pumped by *R*-branch lines of a pulsed HF laser. The observed gains are large (>10% cm⁻¹) and the conversion efficiencies are high. In a ring cavity the system shows directional gain anisotropy characteristic of unidirectional laser amplifiers proposed recently.

In a previous publication¹ very high gains were reported for rotational lines in the first vibrational state of HF gas pumped by the $P_1(J)$ lines² of a pulsed HF laser. After improving the system, some of these rotational lines could also be obtained by pumping with the weaker $R_1(J)$ lines² at 2.4 μ m. As can be seen from the level scheme (Fig. 1), pumping with R lines also opens the possibility of obtaining gain for vibrational P lines at 2.6 μ m. Estimates showed that these gains would also be substantial. This was found to be true.

The HF pin laser used for pumping and the HF gas handling system were identical to those described in Ref. 1. A major improvement in sensitivity was achieved by enclosing the pump laser and the associated electrical equipment in a Faraday cage with rf interference filters. The Monel sample cell was 12 cm long with a 12-mm bore and had sapphire windows placed at Brewsters angle. Sapphire is inert to HF, but, because of the birefringence of this material, care had to be taken to properly orient the optical axes of the windows with respect to the polarization direction of the pump laser beam. The pressure in the cell was determined from the linear attenuation of the pump beam, taking care not to saturate the low-pressure HF gas. The Rand P-branch lines were detected by a fast gold-doped germanium detector cooled to liquid-nitrogen temperature. To distinguish between lines, a monochromator



FIG. 1. HF rotational-vibrational energy level diagram showing R- and P-branch transitions.

with a 16- μ m grating was used.

The pump laser pulses had a duration of from 0.2 to 1.5 μ sec [Fig. 2(a)] and peak powers of 1-2 kW, as measured with an Eppley thermopile. The $R_1(2)$ and $R_1(3)$ lines were strongest and had the longest pulses. The frequency characteristics of pin lasers are known to be poor, ³ possibly with mode jumping and frequency chirping. Spectrum analyzer measurements indicate that our pump laser pulses had a broad frequency spectrum extending over 80-100 MHz. Furthermore, the saturation of the pump transition, as determined from the Rline transmission through the sample cell, is much less than expected if the measured power were contained in a single mode. These findings are important in comparing experiments with theory. Attempts have been made to narrow the frequency spectrum by inserting apertures into the cavity, but the subsequent reduction in pump power brought the system below threshold for pumping.

Laser oscillation was obtained in the $R_1(J) - P_1(J+2)$ three-level systems, J = 1 - 4 (Table I). All of these laser lines have been obtained previously with chemical and electrical-discharge lasers.⁴ Theory indicates that the J=0 system cannot be brought into gain by optical pumping. With sufficiently large pump powers, other systems with J > 5 should also exhibit sizable gains.

First observations were made in a linear cavity of 60cm length, having a 1-m-radius-of-curvature goldcoated mirror and a dielectric-coated mirror with approximately 85% transmission at 2.4 μ m and 95% reflectivity at 2.6-2.7 μ m. The pump radiation was coupled in through the latter mirror, and the $P_1(J)$ lines

TABLE I. Optically pumped HF vibrational transitions.

Pump transition		Laser transition	
Designation ^a	Wavelength µm	Designation ^b	Wavelength µm
$R_1(1)$	2.48	P ₁ (3)	2,61
$R_{1}(2)$	2.45	$P_{1}(4)$	2.64
$R_{1}(3)$	2.43	$P_{1}(5)$	2.67
$R_{1}^{-}(4)$	2.41	P ₁ (6)	2.71

^a $R_1(J)$ signifies the $(v=1, J+1) \rightarrow (v=0, J)$ transition. ^b $P_1(J)$ signifies the $(v=1, J-1) \rightarrow (v=0, J)$ transition.

were coupled out through a 1.5-mm-diam hole in the gold mirror. To reduce losses, the coupling hole was aligned off the beam axis. The pump beam had a diameter of approximately 4 mm, which is about twice the diameter of the lowest-order axial mode of the cavity.

In this configuration all lines oscillated easily. It was noted that for every pump pulse there was a $2.6-\mu m$ pulse, independent of length tuning of the cavity. This indicates a broad gain region for the $2.6-\mu m$ transitions, since the axial mode spacing (250 MHz) is comparable to the Doppler widths of the transitions involved (~ 320 MHz FWHM). This behavior reflects the broad frequency spectrum of the pump laser, the high gain at $2.6 \ \mu m$, and also the fact that the cavity Q is not very high (~ 20% loss per pass).

The output of the linear cavity at 2.6 μ m was very sensitive to small misalignment of the mirrors. This is expected, since the pump beam defines a narrow pencil of gain in the otherwise heavily absorbing unexcited HF gas. To minimize losses it is essential that the cavity mode, defined by the mirror orientation, and the gain volume overlap completely.

To study the properties of the system in detail, a ring cavity was set up [Fig. 3(a)]. As with the linear cavity, every pump pulse produced a pulse at 2.6 μ m, independent of length tuning, and again the output was very sen-



FIG. 2. Oscilloscope traces of pulse shapes; 200 nsec/div. (a) $R_1(2)$ pump laser pulse. (b) $P_1(4)$ output pulse of optically pumped ring laser in forward direction. Note delay with respect to pump pulse.



(b)

FIG. 3. (a) Experimental setup for the study of forward emission only. (b) Ring cavity and coupling scheme to observe forward and backward emission. By blocking (i) [(ii)], backward (forward) pulses can be studied separately.

sitive to angular orientation of the cavity mirrors. A ring cavity is advantageous for optical pumping experiments of this type because there are no reflecting surfaces perpendicular to the pump beam; hence all feedback to the pump laser is eliminated. Furthermore, a ring resonator decouples waves traveling in opposite directions, thus allowing observation of unidirectional effects.⁵

The ring cavity contained the same dielectric mirror described earlier, a 1 -m-radius-of-curvature gold-coated mirror, and a gold-coated flat. The cavity length was 120 cm, again giving an axial mode spacing of 250 MHz. A Corning No. 7905 filter, used as a beam splitter, coupled out approximately 10% of the power. Total losses of this cavity were estimated to be 20%.

As anticipated, ^{1,5} there was a large difference between P-line emission in a direction parallel ("forward signals") and antiparallel ("backward signals") to the propagation direction of the traveling-wave pump radiation, with the forward signals predominating. These features will be described in detail below; the following observations refer to the forward signals only.

The pressures at which signals were observed ranged from about 10-20 mTorr to 200-300 mTorr, depending

on the line studied. At the high-pressure limit the output pulse was a single spike approximately 50 nsec wide. As the pressure was lowered, more spikes (typically three to four) appeared and their size increased considerably. At still lower pressures, the output power peaked and the pulse shape became broad and irregular [Fig. 2(b)]. In this regime the 2.6- μ m pulse began approximately 200 nsec after the start of the pump pulse and lasted until the end of the pump pulse. This is consistent with buildup times estimated from the measured gains of the lines and the cavity Q. Upon further decrease of pressure, the intensity of the pulses decreased again and the shape changed to multiple spikes and then finally to single spikes, until at very low pressures the signals disappeared. At the same time the delay increased until it became equal to the pump pulse length.

The number of molecules available and the extent of saturation set the lower and upper pressure thresholds, respectively, and they determine an optimum pressure for maximum conversion efficiency. These optimum pressures were of the order of 30-50 mTorr for $P_1(3)$ - $P_1(5)$ and about 100 mTorr for $P_1(6)$. This difference in pressures is due to the difference in the ground-state level populations. The saturated absorption in the sample cell ranged from almost total absorption at the high-

pressure limit to almost total transmission at the lowpressure threshold. At optimum output powers, it was typically 40-70%.

The conversion efficiency of the $R_1(2) - P_1(4)$ system was estimated by comparing the size of the signals at 2.4 and 2.6 μ m coupled out by the beam splitter in the cavity. At optimum pressure, where two-thirds of the pump power was absorbed in the sample cell, the transmitted $R_1(2)$ signal and the $P_1(4)$ pulses had about the same peak height. This indicates that about 50% of the absorbed pump power was converted to $P_1(4)$. The actual energy conversion efficiency is smaller since the 2.6- μ m pulses were shorter by approximately 150-200 nsec than the 2.6- μ m radiation in the cavity. The energy conversion efficiency can be quite large, since the frequency differences are small for corresponding R and P transitions (Table I).

To measure the peak gains of the lines at optimum operating conditions, calibrated attenuators were inserted into the cavity. Equating total losses (empty cavity and attenuator) with gain near threshold, the gains were found to be in excess of 10% cm⁻¹. The pump lines for $P_1(4)$ and $P_1(5)$ were strongest, and these two P lines showed gains of 25 and 20% cm⁻¹, respectively. These numbers are quite impressive in view of the low HF pressure in the sample cell.

Gain estimates were obtained from an analysis of threelevel systems by Feldman and Feld, ⁶ extended to include level degeneracy.⁷ This is important for the HF system where certain M_J states in the excited level that are not pumped (selection rules) nevertheless contribute to absorption at the coupled transition, thus reducing the effective gain. The theory⁶ takes into account coherent effects such as two-quantum Raman transitions, and gives higher gains than predictions based on population changes alone. The importance of these contributions to gain is confirmed by laser oscillations in the $R_1(1)$ - $P_1(3)$ HF system, where population considerations alone predict loss even for complete saturation of the pump transition. It is interesting to note that this system has gain in the absence of a population inversion. This feature is also characteristic of lasers pumped by the stimulated Raman effect.⁸

A detailed comparison of theory and experiment, however, is not possible, because the theory assumes a monochromatic pump field, in contrast to the broad frequency output of our pump laser. Estimating the effective power per pump laser mode to be a few hundred W/cm^2 , reasonable agreement is obtained with respect to the magnitude of the gain and the HF pressure at which maximal gain is observed. This agreement also extends to the high-pressure threshold of oscillation and to the saturation of the pump transition as mentioned earlier.

When pumping with a traveling wave, the Raman-type processes⁹ also introduce an asymmetry in gain between waves traveling in a direction parallel (forward) and antiparallel (backward) to that of the pump laser. Be-cause of such coherent effects, the gain in the forward direction is increased and the gain in the backward

direction is decreased, compared to estimates based on population changes alone. This also is included in the analyses of Refs. 6 and 7. For the HF transitions discussed here, substantial gain is expected in the forward direction, but only small gain, or attenuation, is expected in the backward direction. This is one feature of unidirectional laser amplifiers predicted recently.⁵ Another important feature of such amplifiers is the fact that, because of the Doppler effect, gain for forward and backward traveling waves occurs at different frequencies if the monochromatic pump laser is detuned from its line center. Unfortunately, observation of this effect was prevented by the broad frequency spectrum of our pump laser.

To investigate the gain anisotropy the experimental setup of Fig. 3(b) was used. This allowed separate study of forward and backward emission by blocking the beams at (i) or (ii), respectively, and it prevented unwanted feedback between forward and backward radiation. For some of the experiments a different setup was used which allowed simultaneous study of both forward and backward emission. Care was also taken to ascertain that no P-line "superradiance" from the pump laser was present, which could preferentially trigger forward emission.

Compared to the strong forward emission, the backward emission was about 10-20 times weaker and it appeared much less frequently, with one backward pulse per 10-50 pump pulses being typical. Also, the pulse shapes differed from those in the forward direction. Over the whole pressure range the backward emission came in short single pulses of 40- to 60-nsec width, which had about the same delays with respect to the beginning of the pump pulse as the forward emission. Weak backward pulses were delayed by about 50-100 nsec more.

The backward signals had approximately the same lowand high-pressure thresholds as the forward emission. Upon introducing absorbing filters into the cavity to measure gain, it was found that, when backward signals occurred at all, they had only slightly less gain than the forward signals. This is contrary to expectations and will be discussed below.

To further study the backward gain, a small fraction of the forward pulse at 2.6 μ m was reflected into the backward direction. The output of the ring cavity in the backward direction then became very regular, with one pulse for every pump pulse. These pulses had the typical broad or multispiked shape of forward pulses, in contrast to the single spikes usually observed in the backward direction. This shows that waves having the frequency of the forward emission can be amplified in the backward direction, and that some backward gain is always present, despite the fact that laser oscillation in the backward direction occurs infrequently. It also indicates that backward gain occurs over a similarly broad frequency range as the forward gain.

The amplification of the forward signals in these experiments was not uniform. Usually the pulses were amplified by a factor of 8-10, but occasionally the amplification was by a factor of 100 or more. The frequency of

occurrence of this anomalously high amplification was approximately equal to that of normal backward signals (i.e., in the absence of feedback of forward radiation into the backward direction). This suggests that the normal backward pulses occurred under these anomalous gain conditions, whereas the usual backward gain is much less and below oscillation threshold. The observation of high gain in the backward direction is not well understood at present, but it is probably linked to the bad frequency behavior of the pump laser output.

To further test the gain asymmetry, xenon was added to the low-pressure HF gas in the sample cell. At buffer gas pressures above a few Torr the backward signals increased in size and appeared more frequently, while the forward signals were reduced only slightly. Around 50 Torr, forward and backward signals of about equal intensity were observed together for every pump pulse. This can be understood by noting that dephasing collisions tend to reduce the influence of Raman-type processes, which increase the forward gain and decrease the backward gain, as stated above. These observations support the interpretation of the gain asymmetry as being due to coherent effects, and not to some extraneous asymmetry in the experimental setup. At still higher buffer gas pressures laser action occurred only sporadically, because of the decrease in gain caused by collisional deexcitation of the upper level, which becomes dominant.

In conclusion, it can be said that the HF system described shows very definite directional anisotropy of gain as predicted by theory, but, because of the frequency impurity of the pump laser, no quantitative comparison between experiment and theory can be made. A new design for the pump laser, expected to have better frequency characteristics, is presently under construction. The minimum pump powers necessary to obtain oscillation on the coupled transition were estimated to be on the order of 50-100 W in a 4-mm-diam beam for the $P_1(4)$ and $P_1(5)$ transitions. In view of these low threshold powers, cw operation appears feasible.

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Mode-locked high-pressure waveguide CO₂ laser

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Active mode locking of a high-pressure cw waveguide $\rm CO_2$ laser is reported. Pulse widths as short as 3 nsec are obtained. The results are shown to be in good agreement with existing theory.

We have recently shown that by using a hollow dielectric waveguide, ^{1,2} both to guide the laser light and to confine the gas discharge, it is possible to construct small CO_2 lasers with high efficiency and high gain per unit length. ^{3,4} Such a laser is also described in Ref. 5. These lasers have been operated cw at filling pressures as high as 0.5 atm.³ The large gain bandwidth which results from the high-pressure operation allows one to make a single-frequency laser oscillator with a large tuning range, and to make a mode-locked laser with narrow pulse widths. In this letter we describe our experiments investigating the latter possibility by using

an internal acoustic loss modulator to mode lock a high-pressure cw waveguide CO_2 laser.

The experimental setup is shown in Fig. 1. The laser tube consisted of a 10-cm-long 1-mm-diam beryllia (BeO) waveguide tube to which were attached potassium chloride Brewsters-angle windows. Provisions were made for flowing a mixture of $CO_2:N_2$: He through the capillary discharge region, and the bore could be cooled by a coolant flowing through a jacket surrounding the bore. With a simple two-mirror resonator, this tube has produced an output of 2.4 W at 10 μ m when