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[2]For a ruby laser of wavelength $\lambda_{LR}=694.3$ nm, the cutoff density $n_c=2.3\times10^{21}$ cm^{-3}, whereas for a Nd^{3+}:glass laser $\lambda_{LG}=1060$ nm and $n_c=10^{21}$ cm^{-3}.
[3]TEA is defined as transverse excitation atmospheric; see A.J. Beaulieu, Appl. Phys. Letters 16, 504 (1970); V. Merchant and J.C. Irwin, Rev. Sci. Instr. 42, 1437 (1971). A general CO_2 laser bibliography can be found in A.M. Robinson and D.C. Johnson, IEEE J. Quantum Electron. QE-6, 590 (1970).
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[14]For this particular case only 8% of the laser energy is adsorbed. Of this 8%, by the end of the pulse 52% goes into increasing the temperature, 40% into the expansion energy and less than 8% into thermal conductivity losses. In the case of much stronger heating (i.e., laser energy $\gg$ 0.2 J), thermal conductivity losses play a much more important role.

Anisotropic Ultrahigh Gain Emission Observed in Rotational Transitions in Optically Pumped HF Gas

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Gain and laser oscillations are obtained on rotational transitions of the first excited vibrational state of HF gas at room temperature, resonantly pumped by the 2.7-μ lines of a pulsed HF laser. Pumping the *P*-branch transitions connecting the ground and first excited vibrational states produces gain at the coupled rotational transitions at 36, 42, 51, 63, 84, and 126 μ. The gain exhibits directional properties characteristic of a unidirectional amplifier predicted by a recent theory. The incremental gains of these lines are very large, in excess of 1/cm, and the lines oscillate easily without mirrors ("superradiance").

This letter reports observation of gain and laser oscillations in rotational transitions of the first excited vibrational state of pure HF gas at room temperature, pumped by the 2.7-μ lines of a pulsed HF laser. The gain shows anisotropy characteristic of a unidirectional amplifier predicted by a recent theory.[1] The gain in these lines is enormous, and they oscillate easily without mirrors ("superradiance").

Laser oscillations of rotational lines in excited vibrational molecular states have been produced in other gases by optical pumping from the ground vibrational state.[2] The present study also makes use of optical pumping, but the emitted radiation shows unidirectional features not obtained before. These features are caused by the traveling wave pumping of Doppler-broadened transitions. The ultrahigh gain of these lines in HF facilitates the observation of the effects.

The HF sample cell was pumped in a single pass by one of the 2.7-μ $P_1(J)$ lines of an HF laser, where the notation $P_1(J)$ indicates *P*-branch transitions connecting the ground and first vibrational states of HF: $(v=1, J-1) \rightarrow (v=0, J)$. Gain was observed at the coupled rotational transition $(J-1)\rightarrow(J-2)$ in the excited vibrational state [Fig. 1(a)]. These transitions fall in the far infrared (36–126 μ).

The pump laser was a helical transverse discharge HF "pin" laser using a mixture of SF_6 and H_2.[3,4] The laser cavity consisted of a 2-μ diffraction grating and a gold-coated sapphire flat. Individual $P_1(J)$ laser lines could be selected by tuning the grating. Power was coupled out through a 4.5-mm-diam hole in the gold coating of the flat. The output pulses typically had a length of 1 μsec and a peak power of 3–4 kW, as measured with an Epley thermopile.

The absorption cell and the HF gas-handling system were made of Monel metal. The cell was 12 cm long with an inner diameter of 12 mm, and had silicon Brewster windows. The windows were coated with a thin layer of Halocarbon standard stopcock grease to prevent degradation due to HF corrosion. Each window had a transmission of approximately one-half in the wavelength range of interest.

The HF gas was purified several times by freezing it at liquid nitrogen temperature and then pumping on it with

TABLE I. Wavelengths of far-infrared rotational oscillations for *P*-branch transitions in HF.

Pump transition		Coupled transition ($v=1$ excited state)	
Designation[a]	Wavelength (μ)	$J_{upper}\rightarrow J_{lower}$	Wavelength (μ)
$P_1(3)$	2.608	$2\rightarrow1$	126.5
$P_1(4)$	2.639	$3\rightarrow2$	84.4
$P_1(5)$	2.672	$4\rightarrow3$	63.4
$P_1(6)$	2.707	$5\rightarrow4$	50.8
$P_1(7)$	2.744	$6\rightarrow5$	42.4
$P_1(8)$	2.782	$7\rightarrow6$	36.5

[a]$P_1(J)$ signifies the $(v=1, J-1)\rightarrow(v=0, J)$ transition.

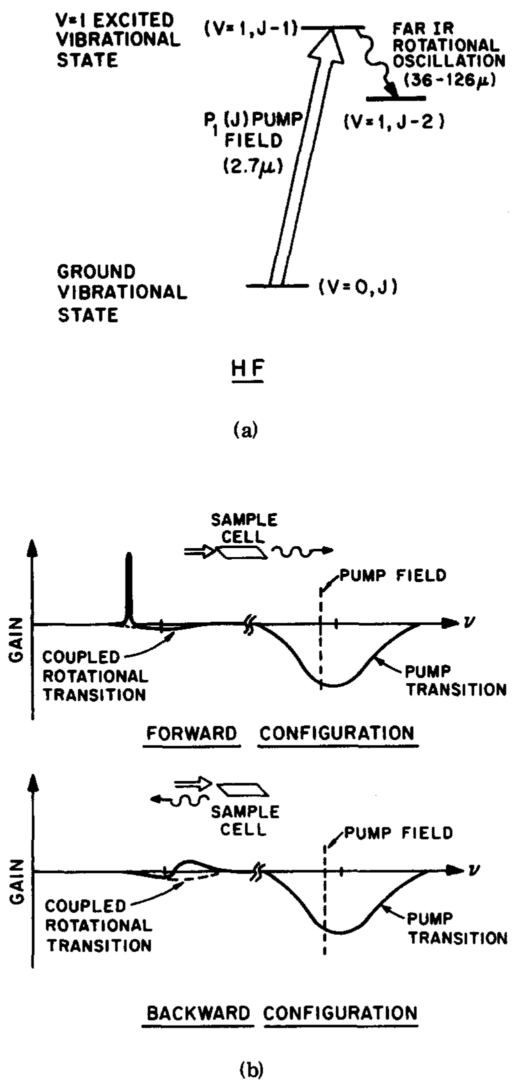

FIG. 1. (a) Energy level diagram. The level (v, J) has vibrational quantum number v and rotational angular momentum quantum number J. (b) Gain profile of an HF rotational transition, resonantly pumped at a coupled vibrational transition. The gain in the forward and backward directions occurs over narrow frequency intervals symmetrically located about line center. Note that the gain in the forward direction is larger than that in the backward direction.

a diffusion pump. To eliminate water vapor contamination, the HF was kept in a Monel cold trap maintained at -30 to $-40\,^{\circ}$C by an ethyl-alcohol—water slush. The absorption cell was filled with HF to an appropriate pressure and then isolated from the rest of the system. The pressure was ascertained from the linear attenuation of the pump beam, suitably attenuated using filters. Pressures ranged from 50 mTorr to 6 Torr, depending upon the line studied.

The far-infrared lines were detected with a helium-cooled In-Ge detector. The pump lines were filtered out using a thin sheet of black polyethylene. A monochromator with a 135-μ grating was used to determine the wavelengths of the rotational lines.

The observed rotational lines are given in Table I. None of these lines have been observed before.[5] All of the lines were strong and easily detectable except for the 63.4-μ line, which is strongly absorbed by atmospheric water vapor. The lines all oscillated without external mirrors.[6] This implies very high gains since the absorption cell is only 12 cm long. The output powers were estimated from the detector calibration to be in the milliwatt range.

The intensity of the lines depends strongly on pressure. This is due to a trade-off between the number of atoms available and the extent of saturation. At low pressures the saturation is high but the number of atoms is low, whereas at high pressures the reverse is true. This implies an optimum pressure for a given pump power. For all of the observed lines the optimum pressure was found to be such that approximately 50—75% of the pump beam was absorbed.[7] These pressures ranged from approximately 50 mTorr for the $P_1(3)$ and $P_1(4)$ pump lines to about 6 Torr for the $P_1(8)$ line. This large variation is due to the fact that the absorption coefficients of the pump transitions change by two orders of magnitude going from $P_1(3)$ to $P_1(8)$.

The unidirectional theory[1] predicts larger gain in the forward direction (i.e., parallel to the propagation direction of the 2.7-μ pump field) than in the backward direction. The gain anisotropy was first studied by comparing the intensities of the radiation emitted by the sample cell in the forward and backward directions under the high-gain conditions where regenerative feedback (mirrors) was unnecessary. The intensities in the forward direction were found to be 5—10 times larger than those in the backward direction.[8]

To further investigate the gain asymmetry, we placed the sample cell in a ring cavity[9] consisting of two gold-coated mirrors having 8-m and 1.5-m radii of curvature, respectively, and a BaF_2, KBr, or NaCl flat, depending on the far-infrared line studied (Fig 2). The pump power was coupled in through the flat, which is transparent in the near infrared and highly reflective and also opaque (Reststrahlen bands) in the far infrared, thus serving to decouple the gain cell from the optics associated with the pump laser. The cavity was about 1 m long. About 1% of the far-infrared power was coupled out by means of a Saran Wrap beam splitter. The

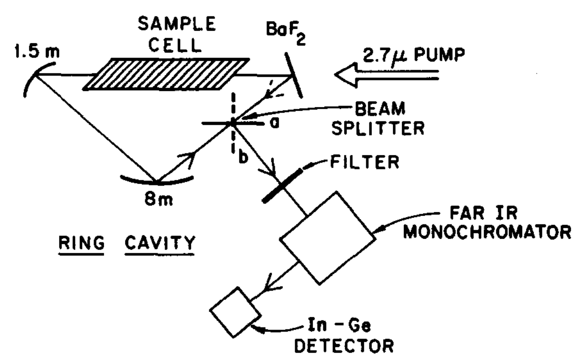

FIG. 2. Ring laser cavity configuration. Forward (backward) far-infrared output power is coupled out of the cavity when the beam splitter is in position a (b).

forward and backward intensities were studied separately. In each case the cavity length was tuned to give maximum output power. The maximum power emitted in the forward direction was found to be 40–400 times larger than the maximum backward power. In the ring configuration the intensity was very stable, whereas in an open system the intensity fluctuated strongly from one pulse to the next.

In another test the pump laser intensity was attenuated by a factor of 15 (peak power ~200 W), using absorbing glass plates. Under these conditions the ring cavity oscillated only in a direction parallel to the pump beam. No signal could be detected traveling in the opposite direction. However, the forward signal was close to threshold and not very stable. This is consistent with the theoretical expectations discussed below.

A brief account of the unidirectional effect may be useful. A normally absorbing transition can be brought into the amplifying phase by resonantly pumping a coupled absorbing transition with an intense monochromatic field [Fig. 1(a)]. If both transitions are Doppler broadened, the gain occurs over a narrow frequency interval determined by the homogeneously broadened linewidth (e.g., due to collisions). It was pointed out in Ref. 1 that, if the pump field is in the form of a traveling wave, gain at the coupled transition for waves traveling in the forward direction occurs at one frequency, whereas the gain in the backward direction appears at a different frequency, symmetrically located on the opposite side of the Doppler profile [Fig. 1(b)]. The frequency separation between forward and backward gain regions is proportional to the detuning of the pump field from the line center of the pump transition, and, if the pump frequency is tuned close to the line center, forward and backward gain regions overlap. An important feature of this effect (independent of whether or not the gain regions overlap) is that the gain in the forward direction is larger than that in the backward direction. This asymmetry arises from the well-known width difference between forward and backward change signals observed in laser-induced line-narrowing experiments.[10,11] The difference in gain can be very large. This explanation accounts for the forward-backward intensity asymmetry observed in the experiments described above.

In applying these considerations to HF, an important fact must be noted. Goldhar *et al.*[3] have found that a transversely excited HF laser always oscillates within a range of less than 70 MHz about line center. It is therefore expected that in our experiments the forward and backward gain regions should overlap. To verify this the single-pass output, either forward or backward, was reflected back through the cell by means of a BaF_2 flat placed normal to the pump beam. (BaF_2 was used to prevent the pump laser beam from being reflected back and, hence, producing a standing wave, which would eliminate unidirectional features.) Under these conditions the output intensity was considerably larger and much more stable than in the single-pass configuration. In fact, the double-pass arrangement enabled the observation of signals which could not be detected otherwise. The fact that the output intensities in the single-pass configuration fluctuated considerably from pulse to pulse, even though the output from the pump laser was fairly stable, indicates spurious feedback due, for example, to back scattering from dust particles or windows or backward Rayleigh scattering in the air.[6]

A rough estimate of the gains of these lines can be made from the observation that in the ring cavity configuration, where a gain of about 1/pass is needed to overcome losses, the threshold power is approximately 200 W. Therefore, at normal operating powers (3–4 kW) the gain coefficient should be 15–20 times larger, leading to an incremental gain of over 1/cm. The theoretically predicted values are roughly consistent with these estimates.

Only qualitative agreement between theory[1,12] and experiment can be expected at present, since the theory assumes a monochromatic pump field interacting with fully Doppler-broadened transitions. The frequency purity of pulsed HF "pin" lasers is notoriously poor.[3] Frequency chirping and mode jumping may occur during a pulse due, in part, to changes in the refractive index of the plasma during the discharge pulse. Furthermore, the unidirectional effect is expected to decrease for the shorter-wavelength lines (36–50μ), which oscillate best at sample-cell pressures where the collision-broadened width is comparable to the Doppler width. However, the longer-wavelength transitions are fully Doppler broadened at optimum pressures.

Laser oscillation on one rotational transition can, in principle, produce cascade oscillations on other transitions lower down in the rotational ladder, especially if the laser oscillation saturates the rotational transition. However, our far-infrared lines are too weak to saturate their transitions, and cascading has not been observed.

In our studies $P_1(J)$ lines were used to pump the excited state rotational transitions. The $R_1(J)$ lines near 2.4 μ $[(v, J) = (1, J+1) \rightarrow (0, J)]$ are also produced by our HF laser, but at powers about 10 times smaller. In view of the low thresholds observed, the $R_1(J)$ lines can probably also be used as pump fields. Pumping of the $R_1(J)$ transitions also opens the possibility of observing unidirectional emission of $P_1(J)$ lines near 2.7 μ.

It is interesting to note that for amplification observed at wavelengths longer than 50 μ, kT at room temperature exceeds the quantum energy $h\nu$, hence the output signals are triggered by thermal radiation. An attempt was made to observe this effect by placing a globar heat source at the amplifier input. Unfortunately, spurious feedback as described earlier caused considerable fluctuations in the output signal, thus masking the effect. Further studies are currently under way.

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[6]There are indications that in this configuration nonresonant feedback due to scattering, e.g., from the cell windows or from air, influences the observed output; hence, the assumption of single-pass gain may not be valid. For a discussion of nonresonant feedback, see R.V. Ambartsumyan, N.G. Basov, P.G. Kryukov, and V.S. Letokhov, Sov. Phys. JETP 24, 481 (1967) [Zh. Eksperim. i Teor. Fiz. 51, 724 (1966)]; R.V. Ambartsumyan, S.P. Bazhulin, N.G. Basov, and V.S. Letokhov, Sov. Phys. JETP 31, 234 (1970) [Zh. Eksperim. i Teor. Fiz. 58, 441 (1970)].
[7]Note that this figure refers to saturated absorption, as opposed to the linear absorption used to measure the pressure.
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[9]Note that a ring cavity decouples forward and backward traveling waves and, at the same time, eliminates the question of spatial gain variations since the cell is traversed many times by the far-infrared radiation.
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Orange Laser Emission and Bright Electroluminescence from $In_{1-x}Ga_xP$ Vapor-Grown *p-n* Junctions *

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$In_{1-x}Ga_xP$ laser diodes which emit coherently at 80 °K at wavelengths as short as 6105 Å with threshold current densities of 4000–6000 A/cm^2 have been fabricated from vapor-grown $In_{1-x}Ga_xP$ *p-n* junction structures. This is the shortest-wavelength laser emission and the first example of coherent *orange* emission yet reported from a semiconductor *p-n* junction. From vapor-grown $In_{1-x}Ga_xP$ spontaneous-light-emitting diodes, external quantum efficiencies in excess of 0.1% have been obtained for orange and red emission at room temperature.

The direct nature of the energy-band structure of $In_{1-x}Ga_xP$ for energies as large as 2.2 eV [1,2] offers significant potential in this alloy system for spontaneous and coherent light emission throughout the red, orange, and yellow portions of the visible spectrum. However, the external efficiency of $In_{1-x}Ga_xP$ spontaneous-light-emitting junctions prepared to date has been limited to values of typically $(1-5)\times10^{-4}$ at room temperature, [3,4] which is far less than predicted for this alloy. [5] Only recently has the quality of *p-n* junctions in $In_{1-x}Ga_xP$ become sufficient to provide coherent emission, [6] with lasers reported only in the infrared spectral region and only at relatively high threshold current densities.

To date, most $In_{1-x}Ga_xP$ has been grown from the melt[7,8] or deposited by liquid-phase epitaxy, [9,10] with *p-n* junctions formed subsequently by Zn diffusion. The vapor-phase growth of $In_{1-x}Ga_xP$ may offer advantages over these growth techniques in that such material can be epitaxially deposited as large-area single-crystalline layers of relatively uniform alloy composition. In this letter, we describe the electroluminescence performance of vapor-grown $In_{1-x}Ga_xP$ *p-n* junctions which recently have been prepared[11] with significantly improved electrical and luminescence characteristics over those from previous vapor-grown material. [12] From such junctions, laser diodes have been fabricated which emit coherent radiation with wavelengths as short as 6105 Å ($h\nu = 2.03$ eV) at 80 °K. To our knowledge, this is the shortest-wavelength laser emission and the first example of coherent orange emission yet reported from a semiconductor *p-n* junction. In addition, spontaneous-light-emitting diodes yielding external quantum efficiencies in excess of 0.1% for orange and red emission at room temperature have been fabricated from such structures.

The $In_{1-x}Ga_xP$ layers described here were epitaxially deposited on ⟨100⟩-oriented *n*-type GaP single-crystalline substrates in a vapor-phase growth system which has been described previously. [12] In the present diodes, the $In_{1-x}Ga_xP$ alloy composition was graded (at a rate of about 2% InP/μm) from GaP at the substrate to the desired composition at the *p-n* junction ($x\approx0.57$) so as to reduce the introduction of lattice misfit dislocations. [13] The *p-n* junctions were prepared *in situ*, using Se and Zn as the donor and acceptor impurities, respectively. The epitaxial growth was about 50 μm thick, with the *p-n* junction located 5–10 μm beneath its outermost surface.

An essential step in the fabrication of laser diodes from the vapor-grown $In_{1-x}Ga_xP$ structures is the removal of the GaP substrate (by lapping and polishing) from the epitaxial layer. This is done to avoid carrier freeze-out, which occurs in the GaP upon cooling to 80 °K. After substrate removal, a slight freeze-out was still observed, which is thought to originate in the GaP-rich