Abstract—The results of experiments on a pulsed optically pumped vibrational transition HBr laser are discussed. In these experiments, rotation-vibration transitions in the first overtone band near 2 μm are pumped with an optical parametric oscillator. Lasing is generated in (2, 1) band transitions near 4 μm. The laser performance is governed by gas phase kinematic processes, which redistribute the absorbed pump energy during the evolution of the laser pulse. A maximum 4-μm output energy of 0.85 mJ was obtained with 3.47 mJ of absorbed pump energy.

I. INTRODUCTION

The hydrogen halides have been the subject of many important spectroscopic and kinetic studies. Their small moments of inertia and strong molecular bonds result in characteristically sparse rotational and vibrational energy level structures. For this reason and others, they have been widely exploited as model systems for studying the energetics of relaxation processes [1]–[3]. Their unusually large infrared line intensities and pronounced vibration-rotation interaction effects have provided important tests of fundamental molecular theory [4]. In addition, their large permanent dipole moments result in large pressure broadenings and shifts which are also of fundamental interest [5]. The hydrogen halides have also been used as the gain media for pulsed and CW chemically pumped mid-IR lasers, most notably the high power HF chemical laser [6].

An electrical discharge activated HBr laser [7] was first demonstrated in 1967 using a mixture of H₂ and Br₂. Following the discharge, the laser is chemically pumped by the reaction,

\[ \text{H} + \text{Br}_2 \rightarrow \text{Br} + \text{HBr}(v) \]

where \( v = 1–5 \) are excited vibrational levels. Lasing then occurs on fundamental \( (\Delta v = -1) \) P-branch transitions in the 4.0–4.6-μm wavelength range. Since the initial demonstration, other experimenters [8]–[12] have employed more sophisticated discharge arrangements, a wide variety gas mixtures, and kinetic modeling [13] in order to optimize the performance of the HBr laser. Although the discharge lasers have produced rather large pulse energies, they are inherently bulky and are not particularly efficient. In this paper, we discuss an optically pumped HBr laser, which has the potential of being both very compact and efficient.

A diagram of the energy levels and transitions involved in the HBr optically pumped laser is shown in Fig. 1. Because of the large vibrational level spacings, very little population exists in any of the excited vibrational levels at room temperature. The rotational levels (labeled by quantum number \( J \)) are populated according to Boltzman statistics. When the output of the optical parametric oscillator (OPO) is tuned to an individual rotation-vibration transition in the (2, 0) band near 2 μm, an essentially complete inversion is produced on relatively strong (2, 1) band transitions near 4 μm. Lasing builds up on the small fraction of spontaneous emission photons that happen to fall within the laser resonator. P-branch transitions \( (J = J' + 1) \) tend to dominate over R-branch transitions \( (J = J'-1) \) because of their larger \( A \)-coefficients and more resilient inversions. Following the laser pulse, the remaining excited vibrational levels eventually relax back to \( v = 0 \) through vibration-to-translation (V-T) processes, releasing some heat into the gas.

An important attribute of the optically pumped HBr laser is that its output radiation near 4 μm is quite cleanly transmitted by atmospheric gases, making it suitable for long-range beam propagation. In addition, the (2, 0) pump band in HBr fortuitously falls in the gain region of a Tm:YAG laser, which can be efficiently pumped by diode laser arrays. Having recently been a significant amount of work devoted to diode-pumped Tm:YAG and Tm:Ho:YAG lasers because of their application in coherent laser radar systems [14]. Many of the issues that need to be considered in designing a pump laser for HBr have already been addressed by the laser radar community.

Tm:YAG is now routinely pumped by diode arrays operating near 785 nm. It was discovered early that a self-relaxation mechanism [15] in Tm:YAG provides two upper lasing levels for each diode laser photon, resulting in unusually high efficiencies. In addition, the modest Stark level splittings and appreciable homogeneous linewidths of the Tm³⁺ ion transitions allows for continuous tuning [16] over the 1.9–2.2 μm range, which overlaps essentially all of the low-J lines in the (2, 0) absorption band of HBr. The center of the strongest laser line in Tm:YAG falls at 2.014 μm. HBr has absorptions from well-populated levels at 2.010 μm \((P(3))\) and 2.018 μm \((P(4))\); hence our particular interest in these pump lines.
be used as needed. The isotopic purity of the HBr was purchased commercially and purified in a freeze-pump-thaw apparatus. The OPO infrared source is a commercially purchased system consisting of an injection-seeded Nd:YAG laser pump source, a narrow-line master oscillator, and a two-stage amplifier arrangement. It can generate tunable radiation in the 1.5–4.0-μm range. The pulses are typically of 5–10 ns duration, with energies of several millijoules in a specified frequency bandwidth of <500 MHz. The linearly polarized pump radiation is focussed into the stainless steel HBr laser cell through CaF₂ Brewster windows. The cell length is 21 cm. A dichroic rear mirror, which is highly transmissive around 2 μm and >99% reflective around 4 μm, is used along with a mode matching lens to ensure that the pump beam is efficiently coupled to the lowest order transverse modes of the optical cavity. The rear mirror has a radius of curvature of 5 m. A flat output coupler is used, resulting in a stable half-symmetric resonator configuration. The resonator length is 34 cm. The resulting TEM₀₀ mode cross section is fairly uniform, with a spot diameter of about 3 mm. Output coupler reflectivities of 90, 80, and 70% were tried. All of the results presented in this paper are with the 80% mirror, which provided the largest pulse energies.

A long-pass filter is used to separate the HBr laser pulse from the unabsorbed fraction of the pump pulse that is transmitted by the output coupler. A pyroelectric detector is used to measure the HBr laser energy and a fast (<2-ns rise time) Ge:Au detector is used to monitor the temporal behavior of the laser pulse. A monochromator with a 150-line/mm grating blazed at 4 μm is used to spectrally analyze the HBr laser output. The HBr gas cell is equipped with side viewing ports with CaF₂ windows for observing infrared fluorescence. Short focal length collection optics along with a 1.6-MHz transient detector and a long-pass filter are used for this purpose.

The HBr was purchased commercially and purified in a stainless steel vacuum manifold using the freeze-pump-thaw method. We found that the HBr lecture bottles contained significant amounts of an impurity (probably H₂) that would not condense at liquid nitrogen temperatures. At 21°C, the vapor pressure of HBr is about 335 lb/in². Small quantities of the purified HBr were trapped into a stainless steel ampule to be used as needed. The isotopic purity of the HBr is dictated by the natural abundances of ⁷⁹Br (50.5%) and ⁷¹Br (49.5%). Capacitance manometers are used to monitor the pressure in the vacuum line and the laser cell.

Absorption lines in the HBr (2, 0) spectrum near 2 μm can be located using direct absorption, laser induced fluorescence (LIF), or laser induced amplified spontaneous emission (ASE). LIF is best because of its nonintrusive nature and almost noise-free background. 4 μm ASE can be observed along the pump beam axis when the laser mirrors are removed and the pump pulse is tightly focused into the HBr cell. ASE is most easily produced at pressures around 50 torr. The pulses are essentially coincident in time with the pump pulse. In HBr, the ASE converter is far less efficient than the mirrored laser because the single-pass gain is not high enough to generate saturating fluences.

The large anharmonicity in the vibrational structure of the diatomic hydrides results in unusually large intensities for the overtone transitions. However, appreciable absorption path lengths and absorber densities are still required to efficiently couple a large fraction of the pump pulse energy into the gas. In order to estimate the expected absorption strengths, the Einstein A-coefficients for the (2, 0) band transitions in HBr were calculated. The coefficients were calculated using the work of Bernage and Niay [17] who used their own measurements and those of others [18]–[22] to compute rotationally dependent dipole moment functions for several fundamental and overtone transitions in HBr. The A-coefficients for the (2, 1) and the (2, 0) band transitions have been calculated and are shown in Tables I and II, respectively. The A-coefficients are expressed so that the absorption (or gain) cross section is written,

$$\sigma(\nu) = \frac{\lambda^2 A}{8\pi} g(\nu)$$

and the population inversion density is written,

$$\Delta n = n_2 - \frac{g_2}{g_1} n_1.$$  

$g(\nu)$ is a normalized lineshape factor and λ is the photon wavelength. $n_1$ and $n_2$ are the population densities and $g_1$ and $g_2$ are the statistical weights of the lower and upper levels, respectively. The instantaneous absorption (or stimulated emission) rate per unit volume is $\sigma A n \omega$, where $\omega$ is the photon density. In cases where the magnitude of the inversion can be driven toward zero (saturation) during the pump pulse, the absorption depends on the photon flux and must be treated using the photon transport approach [23]. For a pulse with a Gaussian spatial intensity distribution ($I(r) \propto e^{-2r^2/\omega^2}$), with radius $\omega$, initial energy $E_0$, and normalized frequency distribution $G(\nu)$, the fraction of $E_0$ transmitted by the gas is given by

$$E_{\text{trans}}/E_0 = \int_{-\infty}^{\infty} G(\nu) \left[ \frac{1}{B(\nu) \eta} \int_{0}^{1} \frac{dx}{x} \cdot \ln[1 + e^{\sigma(\nu) \Delta n (e^{\sigma(\nu) \eta x} - 1)}] \right] d\nu$$

where

$$\eta = \frac{2E_0}{\hbar \nu^2} \omega^2$$

and $\beta = 1 + g_2/g_1$. The natural abundances of ⁷⁹Br (50.5%) and ⁷¹Br (49.5%). Capacitance manometers are used to monitor the pressure in the vacuum line and the laser cell.

Fig. 2. Schematic diagram of the optically pumped laser apparatus.
TABLE I

HBr (2, 1) Band Transition Wavelengths and A-Coefficients

<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (μm)</th>
<th>A-Coeff. (sec⁻¹)</th>
<th>Transition</th>
<th>Wavelength (μm)</th>
<th>A-Coeff. (sec⁻¹)</th>
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<td>P(1)</td>
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<td>4.02559</td>
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<td>P(2)</td>
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<td>P(3)</td>
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TABLE II

HBr (2, 0) Band Transition Wavelengths and A-Coefficients

<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (μm)</th>
<th>A-Coeff. (sec⁻¹)</th>
<th>Transition</th>
<th>Wavelength (μm)</th>
<th>A-Coeff. (sec⁻¹)</th>
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<tr>
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<td>P(4)</td>
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<td>P(5)</td>
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<td>P(9)</td>
<td>2.06412</td>
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</table>

η is the number of photons per unit area in the pulse at τ = 0. ℓ is the length of the laser cell. To the extent that collisional relaxation, spontaneous emission, and diffusion can be neglected, the energy absorbed is independent of the pulse duration. Because of the frequency dependence in the cross section, the results of the absorption calculations depend critically on the frequency domain overlap between g(ν) and G(ν). Both distributions are assumed to be peaked at the resonant frequency in (3). In addition, the inversion is assumed to saturate homogeneously (i.e., g(ν) is constant during the excitation process). This is a reasonable assumption at the HBr laser operating pressures of a few tens of torr, since pressure broadening is substantial. The room temperature Doppler width of the HBr (2, 0) band pump transitions is around 205 MHz. From previous measurements on the (1, 0) band [18], [19] and from comparisons with the pressure broadening trends in the fundamental and overtone bands of HCl and HF [5], we expect a FWHM self-broadening coefficient of about 10 MHz per torr for the low-J transitions in the (2, 0) band of HBr.

At room temperature, the most populated rotational levels in HBr are J = 2-4, with the maximum being in J = 3 with 17% of the population of a given isotopomer. The very small fractional change is mass between the 79Br and 81Br isotopes of bromine result in only slight differences in the reduced masses of the two HBr isotopomers. Therefore, the splittings between the rotational and vibrational energy levels are relatively small, and the spectra are nearly overlapped [22], [24]. However, the line splittings in both the (2, 1) and (2, 0) bands are still too large to allow any coupling between the two HBr isotopomers by either the pump or the lasing fields.
II. RESULTS

Lasing has been observed while pumping on both P-branch and R-branch lines for rotational quantum numbers ranging from 0 to 8. A typical oscilloscope trace of the pump and laser pulses is shown in Fig. 3. The build-up time of the laser is a function of the laser gain, which depends on the amount of pump energy absorbed and the intensity of the pump pulse. For a wide range of conditions, the build-up times range from 20–200 ns, which is a sufficient amount of time for collisions to redistribute the excited state population. Collisional relaxation continues during the laser pulse, which can suppress saturation and deplete the inversions on weaker lines.

Fig. 4 is a graph of the absorbed fraction of 2-µm energy versus HBr pressure. The data was taken over a wide variety of pump pulse energies and intensities. The reasonably consistent data provides an indication that we are not operating under pronounced saturation conditions. The scatter in the data is presumably due to pulse to pulse variations in the energy and frequency of the pump pulse. The conditions used in the calculated curve were chosen to mimic the optimum lasing arrangement of a mode matched pump profile, with approximately 10 mJ of input energy. Under these conditions, saturation effects are minimal and the absorption can be explained quite adequately using a simple Beer’s law analysis. The leveling off in the absorption curve at high pressures is due mainly to pressure broadening, which effectively reduces the absorption strength over the bandwidth of the pump pulse.

Although detailed rotational rate constants have not been measured in HBr, we anticipate from studies on HF that rate constants in excess of $1 \times 10^{-10}$ cm$^3$/molec·s should be expected [2]. The known room temperature V-V and V-T rates in HBr are listed in Table III. In the laser described in this paper, the pump pulse duration and the HBr laser build-up times are short, which allows very little time for vibrational relaxation processes to occur, except at high pressures. The reactions involving HBr ($v = 0$) are the most important because, at any given time in the evolution of the laser pulse, the vast majority of the background gas is in the ground vibrational state. The situation could change as we approach cw pumping or use very energetic pump pulses at high pressures. The rapid energy transfer between isotopomers in $v = 1$ may tend to preserve the population inversion in a longer-pulse laser by speeding the depletion of the lower lasing levels. The slow V-T rate for removing the lower lasing level population may preclude cw lasing and ultimately limit the repetition rate in a pulsed system.

Fig. 5 shows a plot of the laser output versus HBr pressure while optically pumping the P(3) line of the (2, 0) band ($J = 3, v = 0 \rightarrow J = 2, v = 2$). The peak pressure of 20–30 torr occurs as the absorption curve in Fig. 4 starts to level off. At higher pressures, the rotational relaxation rate becomes faster, causing the $J = 2$ population to rapidly redistribute to many other rotational levels during the build-up of the laser pulse. In turn, the gain is reduced on individual transitions, which slows the build-up of the laser pulse and allows more time for V-V processes to quench the upper lasing levels. For this reason, the peak in the pressure curve shifts toward higher values as the pump energy is increased. This is encouraging

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Type</th>
<th>Rate Constant (cm$^3$/molec·s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HBr($v=1$) + HBr($v=1$) $\rightarrow$ HBr($v=2$) + HBr($v=0$) $\Delta E=90$ cm$^{-1}$</td>
<td>V-V</td>
<td>$2.9 \times 10^{-12}$</td>
</tr>
<tr>
<td>HBr($v=1$) + HBr($v=0$) $\rightarrow$ HBr($v=0$) + HBr($v=0$)</td>
<td>V-T</td>
<td>$1.8 \times 10^{-14}$</td>
</tr>
<tr>
<td>HBr($v=2$) + HBr($v=0$) $\rightarrow$ HBr($v=1$) + HBr($v=0$)</td>
<td>V-T</td>
<td>$&lt; 3.1 \times 10^{-18}$</td>
</tr>
<tr>
<td>H$^7$Br($v=1$) + H$^9$Br($v=0$) $\rightarrow$ H$^7$Br($v=0$) + H$^9$Br($v=1$) $\Delta E=0.35$ cm$^{-1}$</td>
<td>V-V</td>
<td>$1.5 \times 10^{-11}$</td>
</tr>
</tbody>
</table>

Fig. 3. Temporal profile of a typical a) HBr laser pulse and b) OPO pump pulse. The horizontal scale is 25 ns/div. The (2, 0) band pump line is P(3) and the HBr pressure is 33 torr.

Fig. 4. Fractional absorption on the (2, 0) band P(3) line in a 21-cm cell of HBr; (o) experimental; (solid line) calculated using (3). The calculation was carried out using the following parameters: a Gaussian beam radius of 0.15 cm, a pulse energy of 10 mJ, an OPO linewidth of 500 MHz, and an HBr self-broadening coefficient of 10 MHz/torr.
when one considers the possibility of scaling the laser to high pulse energies.

If we ignore all relaxation effects and assume that we saturate both the pump and laser transitions, then the amount of energy that can be extracted in 4-μm photons at a given pressure is about 30 μJ/cm² per torr of mixed isotope HBr, depending somewhat on which lines are involved. At 20 torr, the TEM₀₀ mode of our laser can yield around 1.5 mJ of 4-μm photons. When only rotational relaxation is considered, it may be possible to store even more energy, since saturation is partially suppressed in the pumping step by the rapid spreading of the population to other rotational levels. However, at the pressures required to delay saturation during our 5-10-ns pumping pulse, pressure broadening reduces the absorption strength and the present pulse energy is too low to excite a large fraction of the HBr population within the laser mode volume. Rotational relaxation during the pumping step does certainly take place and will become increasingly more important with longer duration pump pulses, like those generated with a typical Q-switched Tm:YAG laser [25].

Fig. 6 shows spectrally resolved output traces of the HBr laser at 20 torr. The laser spectrum depends on which rotational level in v = 2 is populated and not on whether the transition is a P- or R-branch. The laser build-up times for the spectra in Fig. 6 were about 25 ns as in Fig. 3. It is clear from Fig. 6(a) that a large fraction of the J = 2 rotational level population was transferred to J = 3 and J = 4 during this time. By comparing the relative strengths of the lines, we can obtain an estimate of a rate constant for the depletion of the J = 2 population (6 × 10⁻¹³ cm³/molec⁻s). At HBr pressures greater than about 20 torr, the strongest laser line is always P(4), even when pumping the J = 0 level (P(1) line). This implies that thermalization occurs rapidly and the P(4) line accumulates the most gain, simply because the J = 3 level has the largest equilibrium population. Furthermore, once the laser builds up, the population transferred to v = 1 also rapidly rotationally thermalizes and the inversion is maintained on the strongest lines. Fig. 6(b) shows the output spectrum when the J = 3 level is pumped. Although rotational relaxation is rapid, the P(4) line is the first to receive spontaneously emitted seed photons and has an even larger gain advantage over the other lines. When the P(4) line builds up, it rapidly transfers population from the upper to the lower lasing level, causing a temporary rotational nonequilibrium in v = 1 and v = 2.

If there are not a large number of photons at other transition frequencies, the population inversion on the strong line can be repumped by rotational relaxation, thereby stealing gain from the other lines.

The efficiency in which the absorbed pump energy E₆₅₃ can be converted into 4-μm laser energy Eₖ₆₅₃ is determined by several factors. In an extreme case where all of the pump energy is stored in one rotational level and lasing occurs on a single line with no rotational relaxation occurring, the maximum energy conversion efficiency is

\[ \frac{E_{\text{lase}}}{E_{\text{abs}}} = \frac{1}{\beta} \frac{\lambda_{\text{pump}}}{\lambda_{\text{lase}}} \]  

(4)

where for the P(4) line is about 27%. A similar value is obtained if lasing occurs on all P-branch lines from a rotationally thermalized upper vibrational level. With only a few strong lines lasing (P(3), P(4), and P(5)), as in the laser described here, an efficiency of only about 14% would be expected if there were no rotational relaxation occurring during the laser pulse. In another extreme case where rotational relaxation is infinitely fast and a single transition continues to lase until it is saturated with both the v = 1 and v = 2 vibrational manifolds remaining thermalized, the efficiency is

\[ \frac{E_{\text{lase}}}{E_{\text{abs}}} = \frac{1}{1 + \frac{g_2 f_s(J)}{g_1 f_s(J)}} \]  

(5)

where \( f_s(J) \) is the equilibrium fraction of the total population in the level \( J \). The subscripts 2 and 1 on the \( f \) are for the upper and lower vibrational levels, respectively. For the
pumped. Thus far, no lasing has been observed on transitions 1 101 required to efficiently absorb the pump energy into our laser to operate on only a few P-branch lines, no matter which line is does enhance its performance.

The maximum measured efficiency we have obtained thus far is 24.4%, with a maximum output pulse energy of 0.85 mJ. The internal losses due to imperfections in the optics and coatings typically amount to only a few percent loss. However, even though the pulse build up times are short, there is some loss due to vibrational relaxation. The most important process is the endothermic V-V relaxation of HB(r(v = 2)) by HB(r = 0). The AE of the reaction is only 90 cm⁻¹, so the reverse rate constant is appreciable. At 20 torr, about 5% of the upper level is quenched before the laser pulse builds up. When we include this loss, it is apparent that the optically pumped HBr laser is already approaching its maximum theoretical efficiency, and that rotational relaxation does enhance its performance.

IV. CONCLUSION

We have demonstrated an optically pumped HBr laser at 4 μm using short-pulse excitation from an OPO. An energy conversion efficiency of 24.4% was obtained, at a maximum output energy of 0.85 mJ. The laser efficiency improves as more pump energy is coupled into the laser resonator. At the optimum pressures of 20–30 torr, the laser tends to operate on only a few P-branch lines, no matter which line is pumped. Thus far, no lasing has been observed on transitions in the (1, 0) band. Because of the appreciable pressures required to efficiently absorb the pump energy into our laser cell, rotational and vibrational relaxation processes play an important role in the overall laser performance.

REFERENCES