

## Analysis of hydrogen halide lasers

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## Analysis of hydrogen halide lasers

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A comparative analysis is made of the optical and energy characteristics of lasers utilizing binary mixtures containing hydrogen halides in which nonresonant vibrational-vibrational ( $VV'$ ) exchange is used to pump vibrations of the emitting molecules. Specific values are obtained for the gain and specific radiation energy achieved in these systems.

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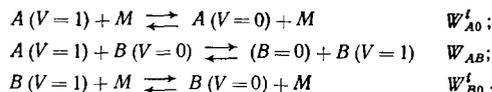
Excitation of metastable levels of diatomic homonuclear molecules followed by energy transfer to molecules with dipole-allowed transitions capable of emitting a coherent energy flux is now widely used to obtain coherent radiation in continuous-flow lasers. This principle has been used to develop both high-power gasdynamic and electric-discharge lasers using  $N_2$  molecules as the energy carrier and  $CO_2$ ,  $N_2O$ , and  $CO$  molecules as the emitter.<sup>1</sup> Of considerable interest from the point of view of their use as the active component are the hydrogen halide molecules: HF, HCl, HBr, HI, DF, DCl, DI, and DBr.

The advisability of using hydrogen halides in mixtures with homonuclear molecules was first noted in Ref. 2. These molecules are of interest since it is possible to obtain a high specific radiation power and highly efficient conversion of thermal or electrical energy into coherent radiation energy.<sup>3</sup> Moreover, the radiation wavelengths of these molecules are within the transparency window of the Earth's atmosphere. All these molecules have an intrinsic dipole moment and thus, when they are used as the active component, high gains can be expected for a small population inversion due to a given vibrational-rotational transition. This is also demonstrated by the results of theoretical studies<sup>4-6</sup> in which the feasibility of developing a  $D_2$ -HCl-He(Ar) gasdynamic laser was considered. A detailed analysis of an  $H_2$ -HF laser using electron-beam excited molecular hydrogen was made in Ref. 3. Earlier investigations of hydrogen halide lasers<sup>7-12</sup> were mainly made for systems where vibrations of the HF, HCl, DF, and HBr emitting molecules were excited chemically or electrically.

The aim of the present investigation was to search for new active media using binary homonuclear molecules and hydrogen halide molecules, to analyze the vibrational energy exchange kinetics in these media, and to estimate the energy characteristics of lasers utilizing these mixtures.

### 1. CHOICE OF ACTIVE MEDIA AND VIBRATIONAL ENERGY EXCHANGE KINETICS IN THESE MEDIA

In order to identify carrier molecules capable of efficiently transferring energy to hydrogen halide molecules, we shall analyze the kinetics of the physical processes in these binary mixtures. In this case, the carrier molecules will be denoted by the letter "A" and the emitting molecules will be denoted by "B". The analysis will be made using the harmonic oscillator model. Relaxation of the energy in each mode can then be considered to be relaxation of a level with the number  $V=1$  and the main energy exchange channels will be as follows:



Here,  $M$  is any mixture molecule ( $M=A, B$ );  $W_{A0}^i$  is the rate of relaxation of the  $V=1$  level in an  $M$  molecule;  $W_{AB}$  is the rate of vibrational-vibrational ( $VV'$ ) exchange between the mixture components.

The equations determining the change in the average number of quanta in the modes in the presence of energy exchange by these channels may be expressed in the form<sup>1</sup>

$$\frac{d\varepsilon_V^A}{dt} = \frac{\rho R}{\mu K} \left[ -\frac{y_A - y_B \exp(-(\Theta_A - \Theta_B)/T)}{(1-y_A)(1-y_B)} W_{AB} \gamma_B - \frac{y_A - y_{A0}}{1-y_A} \sum_{i=1}^n W_{A0}^i \gamma_i \right]; \quad (1)$$

$$\frac{d\varepsilon_V^B}{dt} = \frac{\rho R}{\mu K} \left[ \frac{y_A - y_B \exp(-(\Theta_A - \Theta_B)/T)}{(1-y_A)(1-y_B)} W_{AB} \gamma_A - \frac{y_B - y_{B0}}{1-y_B} \sum_{i=1}^n W_{B0}^i \gamma_i \right]. \quad (2)$$

Here,  $\varepsilon_V^j = r_j y_j / (1 - y_j)$ ;  $r_j$  is the degree of degeneracy of the  $j$ -th mode;  $y_j = \exp(-\theta_j/T_j)$ ;  $y_{j0} = \exp(-\theta_j/T)$ ;  $\theta_j$  and  $T_j$  are the characteristic and local vibrational temperatures of the  $j$ -th mode;  $T$  is the translational temperature of the gas;  $\gamma_i$  is the molar fraction of the  $i$ -th component in the mixture ( $i=A, B$ );  $n=2$ ;  $\mu$  is the molecular weight of the mixture;  $K$  is the Boltzmann constant;  $R$  is the universal gas constant.

The analysis will be made for systems with mixing. At zero time all the energy is concentrated in molecular vibrations of the  $A$  component, i.e.,  $\varepsilon_V^A(t=0)$ , whereas  $\varepsilon_V^B(t=0)$  is governed by its equilibrium value  $\varepsilon_{V0}^B = r_B y_{B0} / (1 - y_{B0})$ . During mixing of the  $A$  and  $B$  components, energy is transferred to vibrations of the  $B$  molecules and maximum pumping of the  $B$  component by the carrier gas  $A$  is governed by the condition

$$d\varepsilon_V^B/dt = 0. \quad (3)$$

Allowing for Eq. (2), we have

$$y_A - y_B \exp(-(\Theta_A - \Theta_B)/T) - (y_B - y_{B0})(1 - y_A) \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A = 0. \quad (4)$$

At gas temperatures  $T = 300$  °K, which will be analyzed subsequently, it can be accurately assumed for hydrogen halide molecules that

$$y_{B0} \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A \ll y_A \ll 1.$$

In this case, we can obtain from Eq. (4)

$$y_B = \frac{y_A}{(1 - y_A) \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A + \exp(-(\Theta_A - \Theta_B)/T)}. \quad (5)$$

It can be seen from Eq. (5) that in order to maximize the relative populations and thus the energies stored in the inverted transition, it is necessary to select the components  $A$  and  $B$  for which there is a large difference between the characteristic temperatures  $\Theta_A - \Theta_B$

and  $W_{AB} \gamma_A \gg \sum_{i=1}^n W_{B0}^i \gamma_i$ .

Mixtures comprising  $H_2$ ,  $D_2$ , and  $N_2$  homonuclear molecules and hydrogen halide molecules satisfying these requirements are given in Table I. Also given are the rate constants  $W_{B0}^i$ ,  $W_{AB}$  at  $T = 300$  °K and  $\sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A$ ,  $\exp[-(\Theta_A - \Theta_B)/T]$  calculated for a mixture with  $\gamma_A = 0.95$  and  $\gamma_B = 0.05$ . The probabilities of the elementary processes were determined on the basis of data from Refs. 12-17 or were calculated using equations given in Refs. 1 and 18.

It can be seen from the results presented in Table I that the following binary mixtures are of the greatest

TABLE I.

Mixture		$\Theta_A$ , °K	$\Theta_B$ , °K	$\Theta_A - \Theta_B$ , °K	$W_{B0}^A$ , cm <sup>3</sup> /sec	$W_{B0}^B$ , cm <sup>3</sup> /sec	$W_{AB}$ , cm <sup>3</sup> /sec	$\sum_{i=1}^n \frac{W_{B0}^i \gamma_i}{W_{AB} \gamma_A}$	$\exp\left(-\frac{\Theta_A - \Theta_B}{T}\right)$
H <sub>2</sub>	HF	5991	5703	288	7.42·10 <sup>-13</sup>	2.03·10 <sup>-13</sup>	1.5·10 <sup>-13</sup>	0.495	0.382
H <sub>2</sub>	HCl	5991	4155	1846	6.10 <sup>-13</sup>	2.10 <sup>-14</sup>	6.10 <sup>-14</sup>	0.1175	0.00216
H <sub>2</sub>	DF	5591	4183	1808	2.25·10 <sup>-13</sup>	7.5·10 <sup>-13</sup>	8.3·10 <sup>-13</sup>	0.319	0.00241
D <sub>2</sub>	HCl	4306	4155	151	6.10 <sup>-17</sup>	2.0·10 <sup>-13</sup>	2.82·10 <sup>-13</sup>	0.000794	0.605
D <sub>2</sub>	DF	4306	4183	123	2.25·10 <sup>-16</sup>	7.5·10 <sup>-13</sup>	8.3·10 <sup>-13</sup>	0.0478	0.645
D <sub>2</sub>	HBr	4306	3685	621	6.2·10 <sup>-16</sup>	2.5·10 <sup>-14</sup>	9.6·10 <sup>-13</sup>	0.00143	0.125
D <sub>2</sub>	DCI	4306	3011	1295	3.18·10 <sup>-16</sup>	7.6·10 <sup>-16</sup>	8.64·10 <sup>-14</sup>	0.0414	0.0135
D <sub>2</sub>	HJ	4306	3325	981	1.17·10 <sup>-16</sup>	9.9·10 <sup>-16</sup>	3.2·10 <sup>-13</sup>	0.0053	0.0392
N <sub>2</sub>	DCI	3357	3011	346	3.06·10 <sup>-16</sup>	7.6·10 <sup>-16</sup>	2.10 <sup>-13</sup>	0.00355	0.315
N <sub>2</sub>	DBr	3357	2649	708	1.75·10 <sup>-16</sup>	5.15·10 <sup>-16</sup>	6.65·10 <sup>-13</sup>	0.303	0.0943
N <sub>2</sub>	HJ	3357	3325	32	3.58·10 <sup>-16</sup>	8.9·10 <sup>-16</sup>	1.95·10 <sup>-13</sup>	0.00445	0.896
N <sub>2</sub>	DJ	3357	2300	1057	1.46·10 <sup>-16</sup>	4.3·10 <sup>-16</sup>	1.15·10 <sup>-14</sup>	0.129	0.0296

interest from the point of view of the criteria specified:  $H_2 - HCl$ ,  $H_2 - DF$ ,  $D_2 - HBr$ ,  $D_2 - DCI$ ,  $D_2 - HI$ ,  $N_2 - DBr$ , and  $N_2 - DI$ .

It should be noted that the use of Eq. (5) may give unjustifiably high values of  $y_B$  (even  $y_B > 1$ ). Values of  $y_B$  and  $y_A$  satisfying the condition  $y_B < 1$  and  $y_A < 1$  may be obtained by using, in addition to Eq. (4), the condition for a constant average number of vibrational quanta in the system before and after mixing:

$$\varepsilon_A \gamma_A + \varepsilon_B \gamma_B = \varepsilon_A(t=0) \gamma_A(t=0). \quad (6)$$

Assuming that  $\gamma_A(t=0) = 1$ , the required system of equations may be expressed in the form

$$y_A - y_B \exp(-(\Theta_A - \Theta_B)/T) - y_B \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A + y_B y_A \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A = 0; \quad (7)$$

$$y_A(1 - y_B) \gamma_A + y_B(1 - y_A) \gamma_A = \varepsilon_A(t=0)(1 - y_A)(1 - y_B). \quad (8)$$

This system of equations is satisfied by

$$y_A = \frac{\varepsilon_A(t=0) - \varepsilon_A(t=0) y_B - y_B \gamma_B}{\gamma_A - y_B \gamma_A - \varepsilon_A(t=0) y_B + \varepsilon_A(t=0)}; \quad (9)$$

$$y_B = B/2 - \sqrt{(B^2 - 4C)/4}, \quad (10)$$

where

$$\varepsilon_A(t=0) \left[ 1 + \exp\left(\frac{\Theta_B - \Theta_A}{T}\right) \right] + \gamma_A \left[ \exp\left(-\frac{\Theta_B - \Theta_A}{T}\right) + \sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A \right] + \gamma_B$$

$$B = \frac{\sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A + \exp\left(-\frac{\Theta_B - \Theta_A}{T}\right) [\gamma_B + \varepsilon_A(t=0) + \gamma_A]}{\varepsilon_A(t=0)}$$

$$C = \frac{\sum_{i=1}^n W_{B0}^i \gamma_i / W_{AB} \gamma_A + \exp\left(-\frac{\Theta_B - \Theta_A}{T}\right) [\gamma_B + \gamma_A + \varepsilon_A(t=0)]}{\varepsilon_A(t=0)}$$

We note that this solution is not quite correct since Eqs. (7) and (8) are contradictory. Equation (8) is the condition for a constant number of vibrational quanta in the system (no dissipation of vibrational energy), whereas Eq. (7) characterizes the equality of the fluxes of vibrational quanta transferred to molecules of the  $B$  component from the carrier gas  $A$  and lost as a result of dissipation due to vibrational-translational exchange. However, since the condition  $l_{VV} < l_{VT}$  is accurately satisfied for all the systems under study, where  $l_{VV}$  and  $l_{VT}$  are the characteristic lengths of vibrational energy exchange between molecules of the  $A$  and  $B$  components and of vibrational-translational relaxation of

the energy of  $B$  molecules, the total quantum losses over the distance  $l_{\nu\nu'}$  are low and it can be considered that condition (8) is approximately satisfied.

## 2. OPTICAL AND ENERGY CHARACTERISTICS OF HYDROGEN HALIDE LASERS

The relationships for  $y_A$  and  $y_B$  derived in Sec. 1 can be used to estimate the gain achieved due the inverted transition and the specific radiation energy stored per unit mass of gas.

We shall use the harmonic oscillator and rigid rotator models to describe the internal structure of hydrogen halide molecules. It should be noted that in diatomic molecules an absolute inversion due to  $V \rightarrow V-1$  vibrational transitions is only achieved by chemical excitation. When other methods of excitation are used, a partial inversion is generally only achieved for  $P$ -branch vibrational-rotational transitions and thus we will subsequently be interested in  $(V, j-1) \rightarrow (V-1, j)$  transitions, where  $V$  and  $j$  are the numbers of the vibrational and rotational levels. Subject to the assumptions made, the gain at the line center of the transition under study may be expressed in the form

$$k_{\nu}^j = \frac{\rho R}{\mu K T} \gamma_B (2j+1) A_{V-1, j}^{V, j-1} \left( \frac{\lambda_0^j}{8\pi} \right)^3 H(a, 0) \sqrt{\mu_B / 2RT} (1-y_B) y_B^{V-1} \delta N; \quad (11)$$

$$\delta N = y_B B_V \exp[-B_V j(j-1)/T] - B_{V-1} \exp[-B_{V-1} j(j+1)/T]. \quad (12)$$

Here,  $\lambda_0^j$  is the emission wavelength at the line center;  $A_{V-1, j}^{V, j-1}$  are the Einstein coefficients;  $H(a, 0)$  is the Voigt function;  $B_V$  is the rotational constant of the vibrational level with the number  $V$ .

In the harmonic oscillator approximation every  $V \rightarrow V-1$  transition with the same number  $j$  emits at the same wavelength  $\lambda_0^j$  and the gain should be calculated as the sum of the contributions  $k_{\nu}^j$  for  $(V, j-1) \rightarrow (V-1, j)$  transitions with  $V=1, 2, \dots, \infty$ . However, as a result of the anharmonicity of the molecules, this situation does not in fact arise so that we shall subsequently be interested in the gain and specific radiation energy for the highest-intensity transition in the harmonic approximation  $(1, j-1) \rightarrow (0, j)$ . For all the molecules under study the value of  $\lambda_0^j$  was determined from

$$\lambda_0^j = \frac{K}{hc} [(\Theta_V - \Theta_{V-1}) + B_V j(j-1) - B_{V-1} j(j+1)]^{-1}. \quad (13)$$

We shall now analyze the Einstein coefficients for hydrogen halide molecules. For the HF, HCl, DF, and DCl molecules these were calculated for various  $V \rightarrow V-1$  transitions up to  $V=15$  in Ref. 19. For the HBr, DBr, HI, and DI molecules the dipole moments in the equilibrium position are known,<sup>20</sup> these being related to the Einstein coefficient, and for the  $nV''j'' \rightarrow mV'j'$  electronic-vibrational-rotational transition these are given by<sup>21</sup>

$$\left. \begin{aligned} A_{mV'j'}^{nV''j''} &= \frac{64\pi^4 \nu_0^3}{3h} \left| \frac{R_{mV'j'}^{nV''j''}}{2j''+1} \right|^2 S_{j'j''}; \\ R_{mV'j'}^{nV''j''} &= \int \psi_n \psi_{V''} \hat{M}_i \psi_{V'} \psi_m \psi_{j'} d\tau_i dr. \end{aligned} \right\} \quad (14)$$

Here,  $\psi_n$  and  $\psi_m$  are the wave functions of the  $n$ -th and  $m$ -th electronic states;  $\psi_{V''}$  and  $\psi_{V'}$  are the wave functions of vibrational states with the numbers  $V''$  and  $V'$ ;

$S_{j''j'}$  is the Hönl-London factor;  $\hat{M}_i$  is the dipole moment operator. In our case, optical transitions take place between vibrational levels of the same electronic state and for the  $P$ -branch transitions under study we have  $S_{j''j'} = j'$ .

In order to estimate the Einstein coefficients of the  $(1, j-1) \rightarrow (0, j)$  transitions in HBr, DBr, HI, and DI, we shall use the harmonic oscillator approximation and we shall also assume that for vibrations of a diatomic molecule the dipole moment depends linearly on  $x$  ( $x$  is the displacement of the molecule from the equilibrium position):  $\hat{M}_i = \mu_i(1 + D\hat{x})$  ( $\mu_i$  is the dipole moment for  $x=0$ ;  $D$  is the proportionality factor).

The wave functions of a one-dimensional harmonic oscillator are well known:<sup>22</sup>

$$\psi_V(x) = \left( \frac{\alpha}{\sqrt{\pi} 2^V V!} \right)^{1/2} H_V(\alpha x) \exp\left(-\frac{\alpha^2 x^2}{2}\right).$$

Here,  $\alpha = 2\pi(M\nu/h)^{1/2}$ ,  $M$  is the reduced mass of the oscillator;  $\nu$  is the normal vibration frequency;  $H_V(\alpha x)$  is a Hermite polynomial of the  $V$ -th degree:  $H_0(\alpha x) = 1$ ,  $H_1(\alpha x) = 2\alpha x$ .

Subject to the assumptions made, the expression for the matrix element of the dipole moment of the  $1 \rightarrow 0$  transition is given by the following integral:

$$R_{10} = \int \left[ \frac{\sqrt{2\pi} 2M\nu}{h} \mu_1 \exp\left(-\frac{4\pi M\nu}{h} x^2\right) x + D \exp\left(-\frac{4\pi M\nu}{h} x^2\right) \right] dx. \quad (15)$$

The integral of the first term in brackets is reduced to the tabulated value by changing the variables  $\alpha x^2 = t$ , where  $a = -4\pi M\nu/h$  and that of the second term is reduced by expressing the function in the integrand as a series near the point  $x=0$ .

We shall assume that for a certain diatomic molecule the Einstein coefficient for the  $(1, j-1) \rightarrow (0, j)$  transition is known and is equal to  $A_m^1$ ; the unknown Einstein coefficient for the other diatomic molecule is then, in accordance with Eqs. (14) and (15), given by:

$$A_0^1 = A_m^1 (\nu_{02}/\nu_{01})^3 (\mu_{12}/\mu_{11})^2. \quad (16)$$

Here, the indices 1 and 2 refer to molecules with known and unknown Einstein coefficients, respectively.

We note that Eq. (16) accurately (to within ~8%) agrees with the results of Ref. 19 where  $A_{mV''}^{nV'}$  was calculated accurately for the HF, HCl, DF, and DCl molecules. Table II gives  $\lambda_0^j(j=1)$  and  $A_0^1$  taken from Ref. 19 or calculated from Eq. (16) for the hydrogen halide molecules under study. Also given are the characteristic vibrational and rotational temperatures and the dipole moments  $\mu_1$ .

An inversion due to the  $(1, j-1) \rightarrow (0, j)$  vibrational-rotational transition is only achieved for those values of  $j$  for which the following condition is satisfied

$$j_{\text{min}} \geq T \ln(y_B)/2 B_0. \quad (17)$$

The radiation energy stored in the inverted transition per unit mass of gas can be determined from

$$\left. \begin{aligned} N^j &= I_{\nu} - I_0^j; \\ I_{\nu} &= R \gamma_B \Theta_B \gamma_B / (1-y_B) \mu; \\ I_0^j &= \frac{R}{\mu} \gamma_B \Theta_B [\exp(2jB_0/T) - 1]^{-1}. \end{aligned} \right\} \quad (18)$$

TABLE II.

Mole- cule	$\Theta_A, ^\circ\text{K}$		$A_0, \mu$	$\mu, 10^{18} \text{ esu}$	$A_0, \text{sec}^{-1}$
	$\Theta_A, ^\circ\text{K}$	$B, ^\circ\text{K}$			
HF	5703	30.2	2.46	1.736	188.6
DF	4183	15.9	3.5	—	54.4
HCl	4155	15.3	3.5	1.07	95
DCl	3011	8	4.8	1.12	9.53
HBr	3685	12.2	3.94	0.788	13.3
DBr	2649	6.2	5.5	0.83	3.85
HI	3325	9.44	4.35	0.382	2.88
DI	2300	4.73	6.25	0.445	0.756

The value of  $l_0^j$  for a given  $j$  decreases as  $T$  decreases and for a given temperature, it decreases as  $j$  increases, so that in order to maximize  $N^j$  it is advantageous to have high values of  $j$ , provided that this is allowed by condition (17) and by the gain  $k_v^j$ .

In order to estimate the efficiency of various laser mixtures, it was assumed that the initial vibrational temperatures of the energy carrier molecules  $T_A$  ( $t=0$ ) are the same, 3000 °K, and that the translational temperature of the stream may be 300, 200, and 100 °K. The pressure of the active medium was taken to be  $p = 0.01$  atm. Table III gives results of calculations of the gain and specific radiation energy in these mixtures for various temperatures of the active medium. Also given are the values of  $j_{\min}$  and  $j$  for which  $k_v^j$  and  $N_j$  ( $j \geq j_{\min}$ ) were calculated. It can be seen from these results that the maximum gain and specific radiation energy are achieved in  $\text{H}_2 - \text{HCl}$ ,  $\text{D}_2 - \text{HBr}$ ,  $\text{D}_2 - \text{DCl}$ ,  $\text{D}_2 - \text{HI}$ , and  $\text{N}_2 - \text{DCl}$  mixtures. On the other hand, in the systems now being studied using  $\text{H}_2 - \text{HF}$  and  $\text{D}_2 - \text{HCl}$  mixtures the values of  $k_v^j$  and  $N^j$  are low (at  $T=300$  °K we find  $k_v^j < 0.1 \text{ m}^{-1}$ ). A  $\text{N}_2 - \text{HI}$  mixture is evidently completely unsuitable as a result of the extremely low values of  $k_v^j$  even at  $T \leq 100$  °K.

It should be noted that in estimates of the specific radiation energy an allowance was only made for the vibrational energy stored in type  $B$  molecules. This is because, although the number of vibrational quanta stored in type  $A$  molecules after transfer of energy to  $B$  emitting molecules is appreciable (this is most typical of mixtures with a low value of  $\Theta_A - \Theta_B$ :  $\text{H}_2 - \text{HF}$ ,  $\text{D}_2 - \text{DF}$ ,  $\text{D}_2 - \text{HCl}$ , and  $\text{N}_2 - \text{HI}$ ), conversion of this energy into coherent radiation is impeded either by the extremely fast vibrational-translational relaxation

TABLE III.

Mixture	$T=300^\circ\text{K}$				$T=200^\circ\text{K}$				$T=100^\circ\text{K}$			
	$j_{\min}$	$j$	$k_v^j, \text{m}^{-1}$	$N^j, \text{J/g}$	$j_{\min}$	$j$	$k_v^j, \text{m}^{-1}$	$N^j, \text{J/g}$	$j_{\min}$	$j$	$k_v^j, \text{m}^{-1}$	$N^j, \text{J/g}$
$\text{H}_2 - \text{HF}$	9	9	0.06	43	6	6	0.4	38	6	6	3.2	88
$\text{H}_2 - \text{HCl}$	9	9	3.4	375	3	5	7.16	770	2	5	4.02	945
$\text{H}_2 - \text{DF}$	9	9	0.5	44	6	6	1.4	36	2	6	0.98	330
$\text{D}_2 - \text{HCl}$	10	10	0.08	33	7	7	0.65	43	4	4	3.9	93
$\text{D}_2 - \text{DF}$	11	11	0.17	49	8	8	0.533	67	5	5	2.55	132
$\text{D}_2 - \text{HBr}$	4	4	4.01	470	2	4	5.44	730	1	4	5.6	950
$\text{D}_2 - \text{DCl}$	4	4	0.9	301	2	4	3.9	731	1	4	4.7	1151
$\text{D}_2 - \text{HI}$	4	4	0.6	282	2	4	1.09	460	1	4	1.5	638
$\text{N}_2 - \text{DCl}$	6	6	1.19	35	4	6	2.54	75	2	6	1.8	125
$\text{N}_2 - \text{DBr}$	7	7	0.831	18.5	5	7	1.487	68	3	7	1.1	3
$\text{N}_2 - \text{HI}$	16	16	0.014	—	11	11	0.0142	0	6	6	0.296	3
$\text{N}_2 - \text{DI}$	6	6	0.172	64	4	6	0.304	104	2	6	0.41	174

TABLE IV.

Mixture	$T=300^\circ\text{K}$			$T=200^\circ\text{K}$			$T=100^\circ\text{K}$		
	$j$	$N^j, \text{J/g}$	$\eta^j, \%$	$j$	$N^j, \text{J/g}$	$\eta^j, \%$	$j$	$N^j, \text{J/g}$	$\eta^j, \%$
$\text{H}_2 - \text{HF}$	—	—	—	6	38	0.96	8	197	4.96
$\text{H}_2 - \text{HCl}$	8	670	16.9	9	910	23	8	1025	25.8
$\text{H}_2 - \text{DF}$	9	44	1.1	8	186	4.7	7	350	8.8
$\text{D}_2 - \text{HCl}$	—	—	—	7	43	1.58	7	198	6.95
$\text{D}_2 - \text{DF}$	—	—	—	8	67	2.4	8	220	7.9
$\text{D}_2 - \text{HBr}$	8	830	29.8	9	965	34.6	8	1050	37.6
$\text{D}_2 - \text{DCl}$	5	500	18	7	1086	39	7	1345	48.2
$\text{D}_2 - \text{HI}$	4	282	10.1	6	565	20.2	6	725	26
$\text{N}_2 - \text{DCl}$	7	50	10.4	8	100	20.8	8	134	28
$\text{N}_2 - \text{DBr}$	7	43	9.15	8	78	16.2	8	107	22.2
$\text{N}_2 - \text{HI}$	—	—	—	—	—	—	—	—	—
$\text{N}_2 - \text{DI}$	—	—	—	5	85	17.7	5	174	36.2

process in the  $B$  molecules or by the high rate of radiative transitions ( $\text{HF}$ ,  $\text{DF}$ , and  $\text{HCl}$  have extremely large Einstein coefficients). These processes result in rapid depopulation of the upper level of the inverted transition and an increase in the gas translational temperature, so that they quench lasing even when appreciable vibrational energy is stored in the carrier molecules. These factors were some of the reasons why a high lasing power was not obtained in an  $\text{H}_2 - \text{HF}$  mixture when  $\text{H}_2$  and  $\text{HF}$  molecules were excited by discharge electrons.<sup>7</sup> An exception is the  $\text{D}_2 - \text{HCl}$  mixture for which the  $VV$  exchange time is much shorter than the time taken for vibrational-translational relaxation of the vibrational energy of the  $\text{HCl}$  molecules. By increasing the pressure of the active medium, the rate of  $VV$  exchange can be increased compared with the rate of radiative decay of the  $V=1$  level during lasing. However, calculations of the characteristics of a  $\text{D}_2 - \text{HCl} - \text{He}$  gasdynamic laser made in Refs. 5 and 6 showed that a specific radiation energy higher than 150 J/g cannot be obtained in this mixture even by cooling the medium to  $T=40-60$  °K.

We shall now analyze the efficiency of conversion of vibrational energy into coherent radiation energy using these binary mixtures. We shall take as the criterion the ratio of the specific radiation energy  $N^j$  to the specific vibrational energy of the  $A$  component at  $t=0$ :  $\eta^j = N^j / l_0^A(t=0)$ . The calculations were made for those values of  $j$  for which, as a result of the  $(1, j-1) - (0, j)$  transition, the gain  $k_v^j > 0.5 \text{ m}^{-1}$  is achieved. Such a value of  $k_v^j$  permits efficient extraction of radiation from the resonator even in a small laboratory system.

The results of calculations of  $N^j$  and  $\eta^j$  at various temperatures of the active medium are presented in Table IV. It can be seen that the highest values of  $\eta^j$  in various mixtures are achieved for different  $T$ . The most favorable mixtures from the energy point of view are  $\text{D}_2 - \text{DCl}$ ,  $\text{D}_2 - \text{HBr}$ ,  $\text{N}_2 - \text{DI}$ ,  $\text{N}_2 - \text{DCl}$ ,  $\text{D}_2 - \text{HI}$ , and  $\text{H}_2 - \text{HCl}$ . In these mixtures at  $T=100$  °K we find  $\eta^j > 0.25$ . Lasers utilizing  $\text{H}_2 - \text{HF}$ ,  $\text{H}_2 - \text{DF}$ ,  $\text{D}_2 - \text{HCl}$ , and  $\text{D}_2 - \text{DF}$  mixtures are less efficient.

In conclusion, we note that the total efficiency of the laser system  $\eta$  is readily determined by knowing the efficiency of excitation of vibrations of the energy carrier molecules  $A$ :  $\eta = \eta_B \eta^j$ . For the  $\text{H}_2$  and  $\text{D}_2$  molecules the value of  $\eta_B$  in the discharge may reach 70% and for  $\text{N}_2$  molecules it can even reach 100% (Ref. 3); for thermal excitation we find  $\eta_B = 10-20\%$ .

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## Low-voltage device for passive mode locking of pulsed infrared lasers

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A reliable mode-locking device which can be used to generate short CO<sub>2</sub> laser pulses has been developed and investigated. The device uses *p*-type germanium filters cooled to 78°K, having a low threshold (10-15 MW/cm<sup>2</sup>) and a short bleaching time (≤0.1 nsec). As a whole, the system can withstand more than 10<sup>7</sup> lasing cycles without appreciable damage to the antireflection coatings and laser elements.

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In the infrared range, mode locking has only been achieved for a CO<sub>2</sub> laser. Gas filters using heated carbon dioxide<sup>2</sup> and SF<sub>6</sub> with added He (Ref. 3), and also uncooled semiconducting filters comprising room-temperature *p*-type germanium<sup>4,5</sup> have been used as nonlinear absorbers. Disadvantages of gas filters are their slow response (~2 nsec) (Ref. 2) and the resonant character of the absorption. In particular, SF<sub>6</sub> can only be used for lasing on the P(20) line of a CO<sub>2</sub> laser (Ref. 3).<sup>1</sup> Compared with other nonlinear filters, *p*-type germanium semiconducting filters have many important advantages including their short relaxation time (~1 psec) and their broad infrared absorption spectrum. In order to avoid high reflection losses, antireflection coatings were applied to the faces of these filters. It was shown experimentally that successful operation of germanium filters at *T* = 300°K requires high optical intensities (10-40 MW/cm<sup>2</sup>) (Ref. 4) close to the damage threshold of the filter coatings and the laser optical elements. This has the result that systems using *p*-Ge filters have poor reliability and a short life

at *T* = 300°K. For example, it was reported in Ref. 5 that a nonlinear *p*-Ge filter could withstand between 10 and 30 lasing cycles at *T* = 300°K. As a result of these disadvantages, neither gas nor semiconducting filters have been widely used up till now.

We proposed a method of mode locking in infrared lasers<sup>5</sup> in which the bleaching threshold of a *p*-type germanium semiconducting nonlinear filter could be reduced by more than an order of magnitude by decreasing the temperature and selecting an appropriate carrier density. A decrease in the bleaching threshold should clearly substantially increase the reliability and life of the total laser system.

It was shown in Ref. 7 that the dependence of the absorption coefficient  $\alpha$  on the optical intensity *I* in *p*-type germanium with a free carrier density of 10<sup>12</sup> - 3 · 10<sup>15</sup> cm<sup>-3</sup> at *T* = 78°K takes the form

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_s}, \quad (1)$$

where  $\alpha_0$  is the absorption coefficient at low optical