Chemical Lasers: HF/DF

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The HF/DF chemical lasers are based on the cold and hot pumping reactions:

\[
\begin{align*}
F + H_2 & \rightarrow HF(v,J) + H & v = 1,2,3 & 34 \text{ kcal mol}^{-1} \ '\text{cold reaction}' \\
H + F_2 & \rightarrow HF(v,J) + H & v = 1 – 8 & 102 \text{ kcal mol}^{-1} \ '\text{hot reaction}' \\
F + D_2 & \rightarrow DF(v,J) + H & v = 1,2,3,4 & 34 \text{ kcal mol}^{-1} \ '\text{cold reaction}' \\
D + F_2 & \rightarrow DF(v,J) + F & v = 2 – 9 & 102 \text{ kcal mol}^{-1} \ '\text{hot reaction}'
\end{align*}
\]

which produce the excited species HF\((v, J)\) or DF\((v, J)\) in a population inversion from which energy is extracted as a laser beam at 2.8 \(\mu\) for HF or 3.8 \(\mu\) for DF. These lasers have been extensively developed since their inception in 1967. Both the continuous wave (cw) and pulsed versions of these lasers were pursued. Since the cw device is the laser of choice for high-power applications, this chapter will concentrate on this type of HF/DF laser.

A schematic diagram of a typical HF/DF laser is shown in Figure 34.1. The HF/DF laser consists of an F-atom generator, a nozzle array to accelerate the F-atom stream and reduce its pressure and temperature, a device to inject H\(_2\) or D\(_2\) and promote their mixing with the F atom stream, an optical resonator to extract the laser beam and a pressure recovery system because these lasers operate at subatmospheric pressures. The power produced by these lasers scales with the flow rates of the reactants: to double the power, the flow rates are doubled.

### 34.1 F-Atom Generators

At the present time, small-scale devices that operate at tens to hundreds of watts are commercially available from Helios Inc. (1822 Sunset Place, Longmont, CO 80501, USA). These lasers use a low-pressure arc to partially dissociate SF\(_6\) to produce...
F atoms. If a high-pressure, high-temperature arc was used, these lasers would produce several kilowatts at the same flow rates. The hundreds of kilowatts to several megawatts lasers react to NF$_3$ with a small amount of D$_2$ or H$_2$ to raise the temperature in the combustor high enough to thermally dissociate the remaining NF$_3$. This results in F atoms, N$_2$ and some DF or HF flowing through the primary nozzles at the exit of which H$_2$ or D$_2$ is mixed with the primary stream to form the lasing species, HF or DF.

### 34.2 Mixing Nozzles

Once the F atoms have been produced, the power of the laser is determined by the effectiveness with which the F atoms and the H$_2$ or D$_2$ are mixed. The physical–chemical characteristics of the HF/DF systems determine the design of the mixing nozzles. The pumping reactions that produce the population inversions are very fast, of the order of microseconds. If the chemicals were premixed, the width of the laser beam would be of the order of a millimetre which results in very large intensities which windows and mirrors could not survive. The exothermicity of the pumping reactions raises the temperature which increases the rates of deactivation of the excited HF/DF. The problems of the heat release and the width of the lasing zone are solved by introducing He and expanding the F atom/He stream through a nozzle. For combustor and high-pressure, high-temperature arc-produced F-atoms, the nozzles must be supersonic nozzles. Typically these will be Mach 5 nozzles which produce 2–10 torr exit pressures and gain zones that are several centimetres long [1]. For low-pressure, high-voltage arc-produced F-atom flows, either supersonic or subsonic nozzles may be used. These nozzles typically produce exit pressures of 2–10 torr and gain zones that are 3–12 mm long. In the supersonic case, they are typically Mach 2 nozzles [2].

The original HF/DF lasers employed alternating primary (F atom) and secondary (H$_2$ or D$_2$) slit nozzles at the exits of which the two streams begin mixing [3]. Because the pressures are so low, the mixing is two-dimensional and diffusive with the mixing layer growing as $x^{1/2}$, where $x$ is the distance downstream measured from the nozzle exit plane. Flow field photography showed that the mixing was slow and that the primary and secondary streams were not fully mixed before the fluid exited the resonator. One of the most successful schemes developed to increase the rate of mixing injected interleaved jets of He near the exits of the primary and secondary nozzles. These jets, called trip jets, caused a rapid increase in the rate of mixing and about a factor of two increase in power. With laser-induced fluorescence, Driscoll [4–6] showed that the trip jets introduced fluid element stretching which dramatically increased the surface area of contact between the primary and secondary streams which increased the rate at which they mixed. Driscoll showed that the same effect could be obtained by putting alternating, interleaved solid ramps at the exits of adjacent nozzles. With this understanding of the fluid dynamic mechanism responsible for the increased mixing due to the side-wall injection in these low-pressure nozzles, the logical development of the trip jet concept led to eliminating the secondary nozzle and injecting the H$_2$ directly into the primary flow near the exit of the nozzle. To shield the H$_2$ from the primary flow until it exited the nozzle, a He jet was placed immediately upstream of the H$_2$ jets. This is illustrated by the TRW HYLTE nozzle [7]; see Figure 34.2.

Another efficient mixing scheme is called the double axisymmetric nozzle. This is a conical hole primary nozzle around the exit of which the H$_2$ or D$_2$ is injected from a circular slit.

![FIGURE 34.2 A schematic diagram of the TRW HYLTE nozzle. (This figure was originally published in Ref. [7], copyright © 1991 by the AIAA, reprinted with permission.)](image-url)
This is illustrated by the Bell Aerospace Textron BCL-13 nozzle, Figure 34.3 [8]. These nozzles are excellent performers.

The choice of nozzle depends upon the mission of the laser. If space and volume constraints do not drive the design, rectangular nozzle banks are usually used. The flow system is linear, with the fluid flowing from the combustor through the nozzle bank to the laser cavity and out through the exhaust system. When space and volume constraints are critical, cylindrical gain generators are used. An example is the TRW Alpha laser which is a prototype for a space-based laser system [9]. In this case, the nozzle bank is wrapped around the surface of a cylinder, see Figure 34.4. The centre of the cylinder is the combustor which produces the F atoms. The fluid flows radially out through the nozzles and the resonator to the exhaust ducts and out to space. The choice of nozzle geometry for the cylindrical gain generator is heavily influenced by manufacturing considerations.

HF/DF laser nozzles typically have throat widths that are 0.10 inch or less. The secondary He and H₂ injection holes are of the order of 0.020 inch diameter spaced several millimetres apart and are staggered on opposite nozzle walls. Since these are low-pressure nozzles, the boundary layers are quite thick.

FIGURE 34.3 A photograph of the face of the Bell Aerospace Textron double axisymmetric BCL-13 nozzle. The primary conical nozzle is surrounded by the conical secondary nozzle. The diameter of the exit of the primary nozzle is of the order of 0.12 inches.

FIGURE 34.4 TRW hypersonic wedge nozzles on the surface of the ALPHA laser cylindrical gain generator. (This figure was originally published in Ref. [9], copyright © by the AIAA, reprinted with permission.)
typically up to 50% of the nozzle exit area. The side-wall injection of the H₂ and He partially blocks the primary nozzle, further reducing its effective area ratio. These effects must be taken into account when designing the nozzle so that the desired exit pressure will be obtained.

With the side-wall injection of the secondary He and H₂, the manifolds for these fluids usually determine the thickness of a nozzle blade. The size of the base area between adjacent primary nozzles may become large enough to provide a recirculation region for ground-state HF/DF, see Figure 34.5. In this case, the base region must be purged with He to prevent the buildup of ground-state HF/DF. If this is not done, up to 50% of the laser power may be lost. The He base purge flow rates are determined experimentally to maximize the power.

34.3 Optical Resonators

Each of the three types of resonators, Fabry–Pérot (two plane, parallel mirrors), stable and unstable, have been used to extract power from the HF/DF chemical laser. Since it generally produces the least power, the Fabry–Pérot resonator is only used for special studies. Normally, the stable or unstable resonator is used to extract power. Since the stable resonator extracts power through a partially transmitting mirror, the stable resonator is usually used when the power is less than tens of kilowatts. At higher power levels, partially transmissive mirrors cannot handle the radiative fluxes that occur. For powers of tens of kilowatts and larger, power is extracted with an unstable resonator which employs all reflective optical elements. The outcoupled beam from an unstable resonator generally has a hole in it so that some of the radiation can be fed back into the resonator to keep the lasing process going. The problem is to design the unstable resonator to produce an outcoupled beam with a uniform phase so that it can be focused to a spot in the far field. Since diffraction effects play a major role in the performance of an unstable resonator, the resonator must be designed with a wave optics code. As a minimum, these models are two-dimensional, and in most cases, a three-dimensional wave optics model is used.

The rotational non-equilibrium kinetic-fluid dynamic model must be coupled with the wave optics model of the unstable resonator or the geometric optics model of the stable resonator. Since the flow and optical axes are perpendicular to each other, the computer models are coupled in an iterative fashion [10]. An initial guess at the intensity distribution on each line is used to run the fluid dynamic-kinetic model to obtain the gain distribution on each line. Then, the optics model propagates the initial guess of the intensity distribution on each line one round trip through the resonator. Each time the optical wave passes through the gain medium, the intensity distribution is modified by the gain distribution from the preceding fluid dynamic-kinetic calculation. After the round trip through the resonator, the new intensity distribution is used in the fluid dynamic-kinetic model to recalculate the gain. When the changes in I and α are less than some small number ε, the procedure has converged. The resulting power, spectra and intensity distributions agree very well with data. With appropriate fluid dynamic, kinetic and optical models, it is possible, in theory at least, to predict laser performance as various aspects of the laser design are changed.

When choosing the outcoupling fraction and the location for the optical axis for an unstable resonator, knowledge of the gain distribution on each of the lasing lines is important. If the optical axis is placed so that the saturated gain distribution of some of the lasing lines does not fill the resonator, a time-dependent oscillation may occur on those lines [11]. The period of the oscillation increases as the magnification of the resonator decreases and the amplitude increases as the fraction of the resonator filled by the line decreases [12]. The oscillations do not occur if the gain medium is strongly coupled to the optical fields diffractively, the Fresnel number \( N_f = (D^2/4\lambda L) < 3 \), or geometrically, the number of round trip...
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passes required for a wave to exit the resonator after leaving the Fresnel core \( n_0 = [\ln(MD_0^2/2(\lambda L)^2)]/\ln M > 3.8 \), where \( D \) is the diameter of the large mirror, \( D_0 \) is the diameter of the small mirror, \( \lambda \) is the wavelength, \( M \) is the resonator magnification and \( L \) is the distance between the mirrors.

Prior to the development of the cylindrical gain generator, HF/DF lasers were rectangular and the optical resonators were developed to extract power from rectangular gain regions. The cylindrical gain region produced by the cylindrical gain generator required the development of an entirely new resonator to extract the power. This resulted in a class of resonators, denoting the high extraction efficiency decentred feedback annular ring resonator (HEXDARR) [13,14]. A schematic view of the HEXDARR resonator is shown in Figure 34.6. Testing of the TRW Alpha laser has demonstrated good performance for the HEXTDARR resonator [15].

For all laser geometries, the mirrors that form the resonator must be protected from the lasant flow. This is accomplished with He purges (jets of He) that are positioned to prevent the laser fluids from entering the mirror cavity. Improperly placed or missing purges will result in ground-state HF/DF building-up in the mirror boxes which reduces the laser power and degrades the mirror coatings. The mirror purge flow rates are experimentally optimized to maximize the power.

34.4 Exhaust Systems

The exhaust system depends upon the application for the laser. For power levels up to tens of kilowatts, blowers backed by mechanical pumps are used. The pump exhaust is scrubbed before discharge to the atmosphere. For hundreds of kilowatts power levels, the laser effluent is pumped with a steam ejector jet pump system [16–18] which also scrubs the exhaust.

34.5 Laser Performance

HF/DF laser performance is characterized by the power of the laser beam and by the spectral content of the laser beam or the power spectral distribution. The power of the laser is determined by the flow rates of the reactants and the effectiveness of the mixing nozzles. The power scales linearly with the flow rates of the reactants. For fixed flow rates, an efficient mixing nozzle may double the power obtained with an inefficient mixing nozzle.

The HF laser operates on several \((v, J)\) transitions (wavelengths) simultaneously, with the spectra generally peaked around \(J=6, 7, 8\) in both \(v=2 \rightarrow 1\) and \(v=1 \rightarrow 0\) vibrational bands. Since rotational relaxation is the fastest collisional deactivation mechanism, the original models of laser performance (e.g. [19–21]) assumed that the HF was in rotational equilibrium. These models were capable of predicting the correct power in each vibrational band. However, they allowed only one \((v, J)\) transition in each vibrational band to lase at a time. They predicted a sequential shift as lasing progressed from low-\(J\) to high-\(J\) lines. In the early 1970s, Polanyi and Woodall, and Polanyi and Sloan performed a comprehensive set of experiments on the hydrogen halides, including HF, that measured the fraction of product molecules that were formed in each \((v, J)\) state for both the cold and hot reactions [22,23]. The data showed that the product molecules were produced in a decidedly non-equilibrium distribution over both rotational and vibrational states. A kinetic model that treats each \((v, J)\) state of the HF molecule as a separate species where the nascent distribution was measured by Polanyi and coworkers was set up. This approach was implemented by Hough and Kerber in 1975 for the pulsed laser and by Sentman in 1975 for the cw laser [24,25]. For the cw case, 21 \(J\) states in \(v=0, 1, 2\), that is 63 states for the lasing molecule, are followed. The results of these models were the prediction of simultaneous lasing on many \((v, J)\) lines in each vibrational band, in agreement with experiment. When the rotational relaxation rate constants in these models are increased by a factor of 10^6 above the measured values, the rotational non-equilibrium models reproduce the results of the rotational equilibrium models.

The computer models still predicted significant power in the higher vibrational bands populated by the hot reaction. Particularly in the HF cw case, under certain conditions some power is observed in the \(3 \rightarrow 2\) band, but generally no power occurs in the \(4 \rightarrow 3, 5 \rightarrow 4\) or \(6 \rightarrow 5\) bands. Bartoszek, Manos

FIGURE 34.6 A schematic view of the HEXDARR resonator used to extract power from the cylindrical gain generator of the ALPHA laser. The dark shaded region is the cylindrical combustor and the light shaded region is the gain zone of the cylindrical laser. (This figure was originally published in Ref. [14], copyright © 1993 by SPIE, reprinted with permission.)
and Polanyi [26] performed a set of chemiluminescence depletion with mass spectrometry experiments that allowed them to measure the relative rates for the deactivation reactions:

\[ \text{HF}(v,J) + D \rightarrow F + HD \text{ and } \rightarrow H + DF \text{ for } v = 3, 4, 5, 6. \]

These measured rates showed that these reactions deactivated the HF \((v=3–6)\) as fast or faster than the pumping reactions produced it. When these reactions were incorporated into the kinetic models, the predictions of which vibrational bands lased were in agreement with data.

Before the collisional decomposition reactions were incorporated into the computer models of the HF laser, these models predicted large increases in performance if the laser could be operated to emphasize the hot reaction by increasing \(F_2\) in the flow [27]. Unfortunately, the experimental data did not come close to the theoretical predictions. When the collisional decomposition reactions are included in the computer models, the predicted performance is in reasonable agreement with the experimental data [28].

The power spectral distribution of the HF laser is primarily determined by the rotational non-equilibrium produced by the pumping reactions and the fact that the lasing process is faster than rotational relaxation which results in many \((\alpha, J)\) transitions lasing simultaneously in each vibrational band.

The essential rotational non-equilibrium kinetics required to model the HF laser are:

**Pumping Reactions**

\[
\begin{align*}
F + H_2 & \rightarrow \text{HF}(1,J) + H \\
F + H_2 & \rightarrow \text{HF}(2,J') + H \\
F + H_2(0) & \Leftrightarrow \text{HF}(3) + H \\
F + H_2(1) & \rightarrow \text{HF}(3) + H \\
F_2 + H & \rightarrow \text{HF}(3) + F
\end{align*}
\]

The \(H_2\) in the first two pumping reactions is \(H_2(0)+H_2(1)\).

**Collisional Deactivation Reactions**

\[
\begin{align*}
\text{HF}(1,J) + M & \Leftrightarrow \text{HF}(0,J'') + M \\
\text{HF}(2,J') + M & \Leftrightarrow \text{HF}(1,J) + M \\
\text{HF}(3) + M & \Leftrightarrow \text{HF}(2,J') + M \\
H_2(1) + H & \Leftrightarrow H_2(0) + H
\end{align*}
\]

where \(M=HF, F, H, H_2, \text{ and DF}\)

**Multiquantum Deactivation Reactions**

\[
\begin{align*}
\text{HF}(2,J') + M & \rightarrow \text{HF}(0,J'') + M \\
\text{HF}(3) + M & \rightarrow \text{HF}(1,J) + M \\
\text{HF}(3) + M & \rightarrow \text{HF}(0,J'') + M
\end{align*}
\]

where \(M=HF, F, \text{ and DF}\)

**Collisional Decomposition Reaction**

\[
H + \text{HF}(3) \rightarrow H_2 + F
\]

**VV Transfer Reactions**

\[
\begin{align*}
\text{H}_2(1) + \text{HF}(2,J') & \Leftrightarrow \text{H}_2(0) + \text{HF}(3) \\
\text{H}_2(1) + \text{HF}(1,J) & \Leftrightarrow \text{H}_2(0) + \text{HF}(2,J') \\
\text{H}_2(1) + \text{HF}(0,J'') & \Leftrightarrow \text{H}_2(0) + \text{HF}(1,J) \\
\text{HF}(1,J) + \text{HF}(2,J') & \Leftrightarrow \text{HF}(0,J'') + \text{HF}(3) \\
\text{HF}(2,J') + \text{HF}(2,J') & \Leftrightarrow \text{HF}(1,J) + \text{HF}(3)
\end{align*}
\]

**Rotational Relaxation Reactions**

\[
\begin{align*}
\text{HF}(2,J') + M & \Leftrightarrow \text{HF}(2,J) + M \\
\text{HF}(1,J) + M & \Leftrightarrow \text{HF}(1,J'') + M \\
\text{HF}(0,J'') + M & \Leftrightarrow \text{HF}(0,J') + M
\end{align*}
\]

where \(M=HF, F, H, H_2, F_2, \text{DF, He}, Q\)

The species denoted by \(Q\) is included to take account of any other combustion or dissociation products that may be present in the mixture and which would contribute to the rotational relaxation but not the collisional deactivation of the lasing species.

### 34.6 HF Overtone Laser

The cw HF overtone laser operates on the first overtone \(\Delta \nu = 2(\nu=2) \rightarrow (\nu=0)\) transitions between 1.3–1.4 \(\mu\) [29,30]. An extensive series of experiments [7,29,31] demonstrated that 60%–90% of the fundamental power is obtainable on the overtone, and the overtone is optimized by the same flow rates as the fundamental and overtone efficiency is independent of mode volume and whether the mixing is slow or fast. Since the overtone is a low gain system, \(\alpha_{\text{OT}} = (1/80)\alpha_{\text{FUND}}\) to suppress fundamental lasing, mirror coatings must be less than 1% reflective over the fundamental wavelengths and highly reflective (99%) over the overtone wavelengths. Optimization of the output power [32] requires careful selection of the reflectivity of the resonator mirrors in terms of their absorption/scattering losses. The overtone mirror design problem led to the development of uncooled silicon optics. Measurements of the fundamental gain while lasing on the overtone \(P_{20}(7), P_{20}(8), P_{20}(9), P_{20}(10)\) [33] showed that lasing on the overtone suppressed the gains of the low \(J\) lines \(P_j(4–6)\) and \(P_j(4–6)\) by 41%–96% and suppressed the gains of the high \(J\) lines \(P_j(7–9)\) and \(P_j(7–9)\) by 3%–44% for a well-saturated overtone laser. The high \(J\) lines are suppressed because their upper or lower levels are directly involved in overtone lasing. The upper levels of the \(P_j(7–9)\) lines are depopulated and the lower levels of the \(P_j(7–9)\) lines are populated by overtone lasing, which decreases their gains. The low \(J\) \(P_j\) lines are suppressed [34] because overtone lasing...
depolarizes the high $J_o=2$ states which blocks the rotational relaxation that populates the low $J_o=2$ states which decreases the gains of the $P_2(4–6)$ lines. The low $J_P$ lines are suppressed because overtone lasing populates the high $J_o=0$ states which increases the rotational relaxation that populates the $v=0$ low $J$ states which decreases the gains of the $P_1(4–6)$ lines.

### 34.7 Line-Selected Performance of the HF Laser

To operate the HF laser in a multiple line-selected mode, a grating is incorporated into the resonator in the off-Littrow orientation [35]. In an unstable resonator, the feedback mirrors are placed at the locations of the positive first-order diffraction peaks of the desired lines. In a stable resonator, one line can be selected in the Littrow orientation and the remaining lines selected by placing mirrors at the locations of the positive first-order diffraction peaks of the desired lines. The possible line combinations are dictated by the grating equation. The grating characteristics, ruling and blaze angle, are selected to permit only first-order diffraction and to ensure that the grating incident angle does not exceed the limit for high grating efficiency [36]. For first-order diffraction peaks of the HF wavelengths of interest, the maximum grating incident angle must be less than 57°. For incident angles greater than this, the grating begins to behave as a grazing optic. Experiments by Chodzko, Gordon et al. and Sentman et al. [37–39] have shown that 55%–80% of the multiline power can be obtained by operating the HF laser in the line-selected mode on two to four lines.

### 34.8 Concluding Remarks

The fundamental processes responsible for the performance, power, spectra and beam quality of cw HF/DF chemical lasers are reasonably well understood. An HF/DF laser can be designed with confidence to meet the requirements of an application. The rotational non-equilibrium distribution produced by the pumping reactions is primarily responsible for the power spectral distribution of these lasers. The basic kinetic processes are fairly well understood. The rate constants for the major kinetic processes are known although some of the energy transfer/redistribution rate constants are less well known. Mixing is the mechanism primarily responsible for the power of the HF/DF laser and the basic fluid dynamic mechanism that controls the mixing is fluid element stretching. Resonator designs that efficiently extract the energy from the gain medium in a beam that can be focused to a spot in the far field have been developed for rectangular and cylindrical gain media. Efficient HF overtone lasing has been demonstrated. The processes responsible for the suppression of the fundamental gain while lasing on the overtone have been identified. The grating characteristics required for efficient line-selected operation of the HF laser have been identified. Line-selected operation of the HF laser has been demonstrated for both stable and unstable resonators.

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