Chapter 1

Introduction

1.1 Introduction

Since 1960, when the first laser emission was reported in the red region of the spectrum from a rod of crystalline ruby, this field has grown almost exponentially [1-2]. Today lasers occupy a special place among most advanced technologies. During the last few decades, Lasers have emerged as the most innovative tool, having wide ranging applications starting from the very common supermarket bar code readers to the highly advanced systems such as nuclear fusion systems for power generation, directed energy weapons. There is hardly any sphere of our life, which is untouched by Lasers. A common man encounters Lasers not only in light shows, but also with the beauticians, eye specialists, orthopedists, to sight a few. In industry lasers are being widely used for material processing, alignment etc. Doctors use it for variety of applications like surgery, dermatology, dentistry, ophthalmology etc. The use of lasers by medical community, industry and by the academic community continues to increase. Many educational institutions are using a wide variety of lasers on regular basis for demonstration of various experiments. These devices produce intense beam of coherent light that can be concentrated to the point where considerable energy densities are required. These could either be pulsed lasers or CW lasers and can produce visible to infrared wavelengths. Pulsed lasers can have energies from microjoules to kilojoules, whereas the CW lasers can have powers ranging from milliwatts to Megawatts [3-5].
Gas lasers such as Carbon dioxide gas dynamic laser (CO$_2$ GDL, \( \lambda = 10.6\mu\text{m} \)), Hydrogen fluoride-Deuterium fluoride (HF-DF, \( \lambda = 2.7-3.4\mu\text{m} \)), and Chemical Oxygen Iodine Laser (COIL, \( \lambda = 1.315\mu\text{m} \)) etc. are infrared gas lasers having wide range of applications in various defense and industrial scenarios [6-9]. The population inversion in GDL is based on basic principle of gas dynamics where hot mixture of gases is rapidly expanded through supersonic nozzle to achieve lasing in the cavity [10-12]. However, in chemical laser the population inversion is based on direct or indirect exothermic chemical reaction taking place in the cavity itself. The chemical laser concept though initially suggested by Polanyi in 1961 [13], but was experimentally demonstrated for the first time in 1965 by Kasper and Pimentel at university of California, Berkeley [14]. The main attraction of chemical lasers is their ability to produce very high powers of the order of megawatts. Most important chemical lasers are listed in table 1.1.

<table>
<thead>
<tr>
<th>Laser</th>
<th>Typical Reactions</th>
<th>Wave length, ( \mu\text{m} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>COIL</td>
<td>( \text{O}_2^+ + \text{I} \rightarrow \text{O}_2 + \text{I}^+ )</td>
<td>1.3</td>
</tr>
</tbody>
</table>
| HF    | \( \text{F} + \text{H}_2 \rightarrow \text{HF}^+ + \text{H} \)  
|       | \( \text{H} + \text{F}_2 \rightarrow \text{HF}^+ + \text{F} \) | 2.6 - 3.5 |
| HCl   | \( \text{H} + \text{Cl}_2 \rightarrow \text{HCl}^+ + \text{Cl} \) | 3.5 - 4.1 |
| DF    | Similar to HF with D substituted | 3.5 - 4.1 |
| H Br  | \( \text{H} + \text{Br}_2 \rightarrow \text{HBr}^+ + \text{Br} \) | 4.0 - 4.7 |
| CO    | \( \text{CS} + \text{O} \rightarrow \text{CO}^+ + \text{S} \) | 4.9 - 5.8 |
| CO$_2$| \( \text{DF}^+ + \text{CO}_2 \rightarrow \text{CO}_2^* + \text{DF} \) (transfer) | 10-11 |

**Table-1.1 Major Chemical Lasers**

Out of all these lasers, HF / DF has been the most studied one because of its applications in various industrial and military scenarios.
The HF/DF laser programs resulted in several multi-100 kW and MW-class systems including BDL, NACL, MIRACL, and Alpha [15-16]. MIRACL was utilized for over 25 years at White Sands Missile Range High Energy Laser Test Facility (HELSTF) for a variety of high-energy laser testing including supersonic missile shoot downs. The Alpha-laser was to be the first Space Based Laser (SBL) demonstrator system, but the program was terminated in 2002 despite having developed and integrated most of the necessary technologies. The system that was closest to military in-the-field development was the ground based DF Tactical High Energy Laser (THEL); this technology was designed as a portable laser for the Army; Mobile Tactical High Energy Laser (MTHEL) and protect against incoming rockets, artillery and mortars (RAM), and had multiple demonstrations of shooting down salvos of incoming rockets.

In 1978, US Navy employed 400 kW DF chemical laser, under United Navy Field Test Program (UNFT) to determine the efficacy of DF laser as weapon. This high power laser weapon destroyed TOW wire-guided anti-tank missiles in flight, using an aiming and tracking system, built by Hughes aircraft, to direct the laser beam on to the fast and small missile. It also destroyed a captive helicopter in 1980. Mobile Army Demonstrator (MAD) planned in 1981, employed a 100 kW DF laser, which was later extended to 1.4 MW, as a prototype for air defence. General Delegation for Armament of France started LATEX laser weapon system program, employing 10 MW DF laser with an advanced aiming system provided by Laserdot. At 10 MW plus output power, it is one of the most powerful military laser systems in the world. Defence Advanced Research Project Agency (DARPA), USA is responsible for the development of 2.2 MW DF lasers based DEW called MIRACL, lasing at 3.8 μm.
HF laser based SBL is a global precision laser weapon in the true sense. It is meant to intercept ICBMs and other strategic and tactical missiles, till catastrophic destruction of the same is achieved. SBL is basically meant to destroy missiles during the boost phase itself. SBL has the capability to intercept the missile in boost phase regardless of the range and can give global protection. Presently, USA is pursuing the SBL-DEW program actively and developing Space based Laser Integrated Flight Experiment (SBL-IFX), as the demonstrator to study various aspects, before launching the actual operational SBL platform. Though there are many significant differences in the design and development of military and industrial laser systems, the approach, however remains the same. The output power levels of military applications are of the order of megawatts, while these are only in tens of kilowatts for industrial laser systems. The range, propagation media and the working environment are the important differences. While the military laser systems have to be rugged and mobile, as they have to work in hostile and harsh environment, the industrial systems are housed in air-conditioned enclosures. On the industrial front, high power lasers are employed for various material processing applications like, drilling, cutting, welding, surface hardening, alloying, cladding, automotive production etc and have emerged as most advanced and sought after technologies [17].

The HF/DF lasers with a wavelength of 2.6–3.4 μm are based on vibrational transition. Their scalability to high power levels enables them to be employed in various defense and industrial systems. Further, it is a "direct" type laser where laser emission is produced by the species generated by a chemical reaction. However, one of the limitations of HF / DF lasers is the toxicity of the constituent elements. In this context, the use of an arc plasma generator is beneficial, since it offers safe decomposition of SF₆ for generation of fluorine atoms to be used subsequently for
lasing action. The dissociation of SF$_6$ is a considerably safer alternative than other options of combustion of F$_2$/NF$_3$ in the H$_2$/D$_2$ environment. Further, plasma heater or generator is a versatile tool for optimizing lasers parameters such as gas flow rate, pressure, temperature, Mach number, etc. In order to optimize the processes and to control the effect generated in arc plasma, it is essential to understand the basic arc discharge phenomena.

Chemical lasers are a part of Gas Dynamic Lasers, which are capable of producing high powers required for practical industrial and military systems. A laser propellant, comprising a suitable mix of chemicals, is combusted or reacted and the exhaust efflux is then directed into an expansion nozzle. The exhaust stream from the expansion nozzle contains highly energetic molecules, which are pumped to an excited state where laser action can occur. If a pair of aligned mirrors is placed on either side of the exhaust stream, laser action will occur as photons bounce between the mirrors, and power can be extracted if one of the mirrors is a partial reflector. While simple in principle, the design and construction of a Chemical Laser is quite complicated. Combustors operate at temperatures as high as 1000 to 2000 deg C, depending on the laser fuel mix used. The expansion nozzles require very precisely controlled flow conditions to work, which results in a complex exhaust system designed to produce the required pressure and flow rates. Some laser fuels and/or their exhaust efflux can be highly corrosive and/or toxic. Mirrors must have very low optical losses, since even a 1 percent loss in a 1 Megawatt laser can result in 10 kilowatts of waste heat dumped into the mirrors.

The first chemical lasers built used carbon monoxide (CO) burned in oxygen-nitrogen, with water being added, to produce the same 10.6 μm band laser action used in carbon dioxide gas discharge lasers. CO burning in N$_2$O and benzene (C$_6$H$_6$)
burning in N₂O were also explored as fuels. While the carbon dioxide laser was the first in this class, the Hydrogen Fluorine (HF), Deuterium Fluorine (DF) and Chemical Oxygen Iodine (COIL) lasers soon followed it. The HF laser uses atomic fluorine and molecular hydrogen to produce 2.7 - 2.9 µm band radiation, using typical fuels such as SF₆ or NF₃, with hydrocarbons used to produce hydrogen. The DF laser, on the other hand, uses ethylene (C₂H₄) burned with a nitrogen trifluoride (NF₃) oxidiser, into which deuterium and helium are injected, to produce 3.6 to 4.2 µm band radiation.

Another significant discovery was the Chemical Oxygen Iodine Laser (COIL), invented by the US Air Force Weapons Laboratory in 1977, and now used in the YAL-1A system. The COIL emits in the 1.315 µm range, and uses chlorine gas and an aqueous mixture of hydrogen peroxide and potassium hydroxide to produce excited oxygen molecules, which react with molecular iodine to produce the laser medium. Conversion efficiencies above 20% were demonstrated very early.

*Since HF / DF lasers are the most important chemical lasers and all the practical systems realized so far have been built around that, the focus of the present work is restricted to these lasers.*

Chemical lasers in general and HF/DF lasers in particular have a few major advantages over the carbon dioxide lasers from efficiency standpoint. Chemical lasers are far more efficient than carbon dioxide laser; typically the output energies in case of HF chemical lasers can be as high as 300 J/g as compared to 40-50 J/g in case of CO₂ GDL. These high efficiencies are important both in battlefield as well as in space applications, where size and weight are at a premium. The shorter wavelength associated with the chemical lasers are extremely advantageous as long as atmospheric absorption is avoided. The smallest spot that can be produced on a target
depends on the laser wavelength divided by the diameter of the output optics. If a 3-4 μm chemical laser were used with same transmitting mirror as a 10.6 μm CO₂ laser, the shorter wavelength laser would produce a focal spot only 30% of the diameter as compared to that of CO₂ laser beam. This would increase the power density on the target by about 10 times making it more effective.

HF lasers have higher efficiencies but the propagation losses are quite appreciable. On the other hand DF laser wavelengths fall in the atmospheric window but the efficiencies are significantly lower; typically 60-65% of those observed in HF lasers. Recently HF lasers have been reported to operate in the overtone mode with output wavelength as 1.33 μm with efficiency as high as 70-90% of the fundamental mode. The development of this type of lasers is because of two reasons, firstly there is a clear atmospheric window and secondly the size of the optics required is much smaller.

In a chemical laser the population inversion is directly produced by an elementary chemical reaction. Most chemical lasers operate on a vibrational rotational transition in a molecule because many exothermic gas phase reactions liberate that energy through a stretching vibration in the newly formed chemical bond. The chemical reactions mostly involve free atoms, since the fuels involved do not react rapidly in their molecular forms.

The basic chemical reaction in case of HF laser is as follows:

\[ F + H_2 \rightarrow HF^* + H + 32 \text{ k cal/mol} \]

\[ HF^* \rightarrow HF + \text{laser} \]

In some cases, where F₂ and H₂ are used for laser action, there exists a chain reaction in which the atomic H reacts with the molecular fluorine in the following manner.
H + F₂ -------- HF⁺ + F + 98 k cal/mol

However, in the case of SF₆ or NF₃ based system, the chain reaction is not possible. Depending on the reaction kinetics, these energies which are liberated in exothermic reactions can produce HF molecules which are excited to very high vibrational levels even upto V = 8. But in the most typical practical conditions, usually V does not exceed 3. In addition, each vibrational level has large number of rotational sub levels associated with it. So as such large numbers of wavelengths are possible. The most commonly studied transitions are V=2 to V=1 to V=0, which corresponds to wavelengths in the range of 2.6 – 2.8 µm. Recently, an overtone transition between V=2 to V=0 has successfully been exploited by suppressing the fundamental transition. This overtone transition corresponds to 1.33 µm. These transitions are shown in the following fig.1.1

![HF Energy Levels](image)

**Fig1.1 HF Energy Levels**

It may be stressed here that fluorine and some of the fluorine-based compounds are highly toxic in nature. For example an exposure of 1 ppm of F₂ for 8 hr is the maximum safe limit. On the other hand compounds such as NF₃ are comparatively
less toxic (typically 10 ppm for 8 hr), yet the handling of these gases may cause health hazards. SF₆ is non-toxic and non-hazardous and that is one of the main reasons that the systems based on SF₆ have been developed and used in the laboratories rather than in isolated areas. Moreover SF₆ is being widely used in electrical industry also as an insulator material, its handling is relatively much easier.

The chemical laser system can be divided conveniently into five regions viz. fluorine atom generator, supersonic nozzle, optical cavity, diffuser and exhaust. Block diagrams of these systems are shown in fig.1.2 and fig.1.3. A brief description of the various subsystems is presented here.

1.1.1 Fluorine Atom Generator

Fluorine atoms can be generated by number of ways such as:

a) DC, RF or microwave discharge in a mixture of F-containing molecules such as SF₆ and a rare gas.

b) Shock wave initiated reaction of F₂O and H₂ diluted with Argon, or a mixture of Argon and Fluorine.

c) Arc heated SF₆

d) Dissociation of F₂ using combustor.

e) Dissociation of NF₃ using combustor.

While the first two techniques have been mainly used for low powers of the order of few watts, arc driven and combustor driven systems have been mainly used for practical system capable of delivering powers in excess of kilowatt level.

In the case of combustion driven system, the combustion of Deuterium or Hydrogen with excess of Fluorine takes place in the reaction chamber. Since these reactions are exothermic in nature, combustor temperatures as high as 4000 °C can be achieved. Usually plenum mixture is diluted with Helium before expanding it through
through a fast supersonic nozzle. To minimize the deactivation processes and the concentration of ground state molecules D₂ is usually added in the combustor during HF lasing and H₂ is used in the combustor during DF lasing. The purpose of the combustor is to contain the hot gases that are needed for the generation of F atoms. If the run times are relatively short, the combustor can be heat sunked and no water-cooling is necessary except to cool the combustor between the runs. Block diagram of typical combustor is shown in the fig.1.2.

![Combustion Driven DF Laser](image)

**Fig1.2 Combustion driven DF Laser**

### 1.1.2 Arc Heater for Generation of F-atoms

Although the HF/DF lasers can be run as purely chemical driven lasers as mentioned in the earlier paragraph, however this type of laser is difficult to use in a normal laboratory environment because of storing problems associated with fluorine etc. Moreover, the availability of deuterium is usually a problem. In order to
overcome these drawbacks and to run the chemical laser in the laboratory, an arc-driven HF laser is an excellent alternative approach. Fluorine atoms are created by dissociation of \( \text{SF}_6 \), injected directly into the arc heater with Helium/ Nitrogen and oxygen as diluents. The use of \( \text{SF}_6 \) as fluorine atoms generator avoid the danger connected with the handling of molecular fluorine. \( \text{SF}_6 \) has an average bond strength of 60 kcal/mol as compared to 35 kcal/mol, that of \( \text{F}_2 \). So the dissociation of \( \text{SF}_6 \) molecules requires higher temperatures usually greater than 2000 k as compared to 1400 k required for the dissociation of \( \text{F}_2 \) molecules.

In the arc – heater system, 80% of the heat loss occurs at anode and rest 20% in the cathode housing. This requires efficient cooling of the anode, which can be realized by circulating the deionized water at high pressure.

The overall performance of the arc-driven laser depends on factors such as the flow rate of \( \text{SF}_6 \), and the nature of diluent. At low \( \text{SF}_6 \) flow rates, \( \text{SF}_6 \) is fully dissociated, and the laser output power is proportional to the \( \text{SF}_6 \) mass flow. At higher \( \text{SF}_6 \) flow rates into plenum, laser-output power decrease. This is largely due to a drop in plenum temperature. As more and more gas is added into the plenum, the temperature decreases, the dissociation of \( \text{SF}_6 \) decreases and thus the concentration of \( \text{F} \) atoms decline.

Helium is generally used in the arc heater system in place of Nitrogen. It offers two major advantages viz. better arc-heater efficiency and larger pressure ratios in the same nozzle. The better efficiency in case of Helium is due to the production of Helium metastable atoms and absence of useless energy stored in the vibrational states of \( \text{N}_2 \). The increased adiabatic expansion ratios are, of course, due to the higher specific heat ratio and the lower molecular mass of Helium as compared to Nitrogen. Typically, the peak power output in case of Helium is almost twice that for Nitrogen.
It may be mentioned that in addition to Helium, Argon is also added in order to prevent the corrosion by the atomic fluorine of the cathode tip, which is made of Tungsten.

The reported experimental results on arc-driven chemical laser system indicate that even under optimum conditions of operation, SF₆ is only 75% dissociated in the plenum. This means that a large amount of Fluorine remains tied up in SF₆, SF₄ and lower dissociation products. Moreover, large quantities of sulfur produced by the arc heater tend to coat all cooled surfaces. To avoid these problems, oxygen is usually added to the plenum assuming that it reacts with the Sulfur and lower Sulfur fluorides, thus increasing the concentration of F atoms thereby reducing the Sulfur-deposition problem.

In HF/ DF chemical lasers, the main operational characteristics of inert diluents are its availability and cost. Since Helium is scarce and expensive, so it is replaced by more readily available and cheaper material like Nitrogen [18]. Although investigations by various researchers show convincingly that a chemical laser with Nitrogen has much poorer energy characteristics than a laser with Helium as mentioned above, but for initial experiments and optimization of laser system Nitrogen is considered as good replacement because of easy availability and very less cost. Fig.1.3 shows the overall configuration of the arc heater along with the other subsystems [19].
Fig. 1.3 Schematic of Arc Driven HF Laser

Typical parameters used, on the basis of the available literature, for the generation of a kilowatt level HF laser with Helium is given below as:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arc power</td>
<td>40 kW</td>
</tr>
<tr>
<td>Plasma Gases</td>
<td></td>
</tr>
<tr>
<td>Ar</td>
<td>0.8 g/s</td>
</tr>
<tr>
<td>He</td>
<td>0.9 g/s</td>
</tr>
<tr>
<td>SF$_6$</td>
<td>2.8 g/s</td>
</tr>
<tr>
<td>O$_2$</td>
<td>0.5 g/s</td>
</tr>
<tr>
<td>Fuel Nozzle</td>
<td></td>
</tr>
<tr>
<td>He</td>
<td>0.5 g/s</td>
</tr>
<tr>
<td>H$_2$</td>
<td>0.7 g/s</td>
</tr>
<tr>
<td>Plenum pressure</td>
<td>1.5 bar</td>
</tr>
<tr>
<td>Cavity</td>
<td>5 torr</td>
</tr>
</tbody>
</table>

1.1.3 Nozzle

The purpose of the cw chemical-laser nozzle is manifold:

a) The rapid expansion freezes the fluorine atom concentration at the high value, which is characteristic of the plenum conditions.
b) The static temperatures and pressure levels are reduced to low values, the low pressure promotes rapid mixing and decreases collisional de-excitation. Moreover, the low translational temperature increases laser gain.

c) The large number of mixing surfaces facilitates the rapid mixing and reaction of the laser reactants, as required so that the laser levels can be pumped faster than they are de-excited by collisions.

In contrast to the nozzles being used for gas dynamic lasers, which actually produce the non-equilibrium vibrational states, the nozzle of the chemical laser generates only a non-equilibrium chemical composition. Because of the long atom recombination times, a non-equilibrium chemical composition can be achieved much more easily and efficiently than vibrational non-equilibrium. The high velocity of supersonic gas jet is an important characteristic of chemical laser. The very rapid deactivation of the excited HF, and therefore, the very short lifetimes of the lasing species, results in short inversion zones. Unless the reacting molecules are transported through the cavity rapidly, diffraction and deactivation losses will dominate the active medium. Most importantly, the nozzles in a chemical laser have to mix the fuel H₂ or D₂, rapidly and thoroughly into supersonic flow.

All these considerations have led to a number of designs of these nozzles. The most commonly used is an array of slit nozzles placed vertically and side-by-side.

1.1.4 Resonator

The optical resonator provides feed back to maintain laser oscillation and also to control the optical phase across the output beam front. Coherent beams have low divergence and an excellent beam quality thereby helping in long distance propagation, focusing beam to maximum intensity etc. Optical feedback system, provided by the resonator is used to improve the spatial and temporal coherence and
to build up resonator modes. In a stable resonator, the curvature of the mirrors are chosen so that an optical ray that wanders away from the optic axis of the resonator is returned to it by focusing action of the mirrors. This greatly reduces the diffraction losses of the resonators due to the lateral leakage of the radiation. For a laser resonator with spherical mirrors of radii of curvature $R_1$ and $R_2$ and the distance between them being $L$, the stability condition is written as

$$O < (1-L/R_1) (1-L/R_2) < 1$$

1.1.5 Diffuser and Pumping System

The spent gases in the optical cavity flow into the diffuser, which converts the low pressure supersonic, flow into a subsonic high-pressure flow. Typical pressure recovery ratios range from 3 to 15. Helium is also sometimes injected along the diffuser sidewalls to energize the boundary layer, which is thickened by the mirror purge gas for improved overall diffuser performance. An efficient diffuser raises the pressure at the inlet to the pump and thereby reduces the volumetric gas flow through the pumping system.

The simplest diffuser used in chemical lasers to date has been a constant area duct of about 50cm length and approximately 5:1 to 10:1 length to height ratio followed by a $6^\circ$ half angle subsonic diffuser. The diffuser inlet mates directly and smoothly to the ends of cavity shrouds. The recovered pressure depends on various parameters such as mass flow rate, temperature, molecular wt. etc.

Two types of pumping systems are generally used in chemical laser systems; ejectors and mechanical vacuum pumping systems. Ejectors that use either steam or Nitrogen as their pumping fluid are suitable for large lasers, whereas the mechanical vacuum pumping systems are preferred in laboratory systems. Typically for kilowatt power level lasers, diffuser is connected with a cylindrical pipe to a $25 \times 10^{-3}$ cm$^3$
vacuum vessel which is evacuated with the roots blower pump of 2000-5000 m³/h pump capacity. It may be mentioned that the gases like HF, H₂ etc. entering the pump are not processed and most of the HF produced by the reaction is absorbed by the oil of the mechanical pumps. The presence of these toxic gases, though does not significantly affect the performance of the pumps, but requires the changing of pump oil more frequently.

1.1.6 Scrubber

HF coming out of the exhaust of the vacuum pump is almost as toxic as F₂, and should be removed before the output of the pumping system is vented to the atmosphere. Scrubbing of the laser exhaust gases is, usually done by passing the gases through a basic solution, which reacts with the hydrogen halides to form watersoluble salts. In a typical scrubber system 3% NaOH is sprayed over polypropylene bed to absorb the toxic gases.

1.2 Literature Survey on HF/DF Chemical Lasers

The studies on HF/DF chemical laser represent some of the most amazing works during the first 40 years in the field of very high-energy lasers, including several systems that made it to the field [20-21]. The HF/DF systems were the backbone of the Reagan-era “Star Wars” program. Gross and Bott [5] in their handbook on Chemical Lasers has given detailed discussions of these systems. Further a summary paper by Sentman [22], a book chapter by Behrens and Lohn [23] that provides a more recent perspective, and Horkovich [15] who outlines several of the HF/DF laser systems.

Most HF/DF chemical laser studies addressed a system characterized exclusively by what was called the “cold reaction,” in which a source of F atoms would mix and react with H₂ or D₂. J. C. Polanyi [13] first proposed the possibility of
the creation of infrared lasers based on the vibrationally excited products of a chemical reaction in 1961 Jerome V. V. Kasper and George C. Pimentel [14] demonstrated a pulsed chemical laser in 1965. First, hydrogen chloride was pumped optically so vigorously that the molecule disassociated and then re-combined, leaving it in an excited state suitable for a laser. Then later on hydrogen fluoride and deuterium fluoride lasers were demonstrated. Pimentel was awarded a patent for a scalable overtone HF laser (United States Patent 4,760,582) in 1971.

The continuous wave (CW) chemical HF laser was first demonstrated in 1969, [24] and subsequently patented by researchers at The Aerospace Corporation in El Segundo, California. This device used the mixing of streams of H₂ and F, within an optical cavity, to create vibrationally excited HF, which lased. The atomic fluorine was provided by dissociation of SF₆ gas using a DC electrical discharge. Based on the initial work on discharge based HF/DF laser, later work at US Army, US Air Force, and US Navy contractor organizations (e.g. TRW) used a combustion approach to provide the atomic fluorine. The latter configuration obviated the need for electrical power and led to the development of high power lasers for military applications.

A "hot reaction" variant, in which atomic hydrogen reacts with fluorine molecules and results in chain reaction, was originally thought to have more potential because of its higher exothermicity, but poor laser performance experiments with the "hot reaction" were found to be the result of a critical collisional decomposition reaction, that decomposed the high vibrational levels of HF/DF into the reactants for the "cold reaction" [25].

Lasing was initially demonstrated on the fundamental Δv=1 bands, and later on the first overtone Δv=2 band of HF by Jeffers [26].
Fundamental lasing $\text{HF (v,J)} \rightarrow \text{HF(v-1,J+1)} + \text{hv} \approx 2.5-3.2 \ \mu\text{m for HF}, \approx 3.5-4.2 \ \mu\text{m for DF}$

Overtone lasing $\text{HF (v,J)} \rightarrow \text{HF(v-2,J+1)} + \text{hv} \approx 1.35-1.40 \ \mu\text{m}, \approx 1.85-2.00 \ \mu\text{m for DF}$

A far more comprehensive and recent review of HF/DF kinetics has been presented by Manke and Hager [27].

Spencer D.J., Mirels, H., Jacobs, T.A., Gross, R.W.E. [24] initially reported their “Preliminary performance of a cw chemical laser” with arc-driven Nitrogen based laser system in 1970. Hydrogen was diffused into a supersonic stream containing F atoms. Population inversion was due to the reaction $\text{H}_2+\text{F} \rightarrow \text{HF(v)+H}, \Delta\text{H}=-31.7 \ \text{kcal/mole}, \ v=1, 2$. An atomic F flow rate of 0.030 moles/sec produced 475 W of laser power in the 3-μm region. This represented 12% of the chemical energy involved in the above reaction. The HF chemical laser was later on realized using an arc-heater and SF$_6$ to provide F atoms [28]. The mixture was expanded to form a supersonic jet into which H$_2$ (or D$_2$) was diffused. Population inversion and lasing were due to the reaction $\text{H}_2+\text{F} = \text{HF (v)+H; v}\leq3 \text{ and } \Delta \text{H}=-31.7 \ \text{kcal}$. Power levels above 1 kW were obtained in a N$_2$ diluent gas flow, and levels above 1.7 kW were obtained in a He diluent gas flow. Specific power yields were 43 and 108 kW sec/lb for the N$_2$ and He systems respectively. The variation of laser power and efficiency with cavity optical axis location and SF$_6$ mass flow were also reported for both N$_2$ and He systems.

Brandelik and Paulson reported the operation of an HF /SF$_6$ + H$_2$/ TE laser at high pulse repetition rates [29]. They have used a flowing HF transverse excitation (TE) discharge laser and could run lasers at 10,000 pps for up to one billion pulses.
Preliminary testing at 100 pps produced 350-microjoule/pulse outputs with reduced outputs at higher pulse rates.

Recently, Bulaev et al have realized a high-power non-chain electric-discharge repetitively pulsed HF laser based on SF₆—hydrocarbon mixtures. They have been able to achieve pulse energy of 67 J at a pulse repetition rate of 20 Hz [30].

AS Bashkin et al [31] have reported that the HF lasers can be fine tuned vibrational-rotational lines. An investigation was made of the energy characteristics of a chemical HF laser utilizing an H₂–SF₆ mixture. Efficient lasing was obtained at gas mixture pressures up to 5 bar, specific output energy of 50 J/liter was obtained at 1.5 bar. They were able to tune continuously the lasing frequency in a range 2.2 cm⁻¹ centered on each vibrational-rotational line and thus build a high power tunable laser of the kind required in laser-chemical investigations.

Davis et al [32] have used microwave source instead of arc heater to generate fluorine atoms for HF lasers. They have reported that the fluorine atoms generated using this method can be used for development for not only HF laser, but also for All Gas Iodine Laser (AGIL): a new variant of COIL system. The system uses high power microwaves to produce high enthalpy plasma that thermally dissociates molecular species such as SF₆ and F₂. Results of the characterization of the flow have also been presented.

Panchenko et al have studied in detail the effect of different excitation parameters on discharge stability in mixtures with SF₆ and efficiency of discharge non-chain HF (DF)-lasers using excitation by inductive and LC-generators [33]. Experimental conditions providing maximal performance of discharge non-chain HF (DF)-lasers have been determined. Uniform electric field in the laser gap, uniform preionisation, relatively short current pulse duration and input energy around 50-70
J/liter provide perfect discharge uniformity and greatly improve electrical efficiency of the discharge HF (DF)-laser. Electrical efficiency of the lasers up to $\eta_0 = 5-6\%$ with the output over 1 J was demonstrated for the first time. The effect of preionisation on volume discharge formation in mixtures of H$_2$ and hydrocarbons with SF$_6$ was also suggested.

The demonstration of the first HF Overtone laser by Jeffers[26] enabled the high energy HF laser to operate at a more attractive wavelength range of 1.35 – 1.40 $\mu$m and set off a new wave of detailed experimental and theoretical studies by different groups. The overtone wavelengths were of significant interest to the military for both atmospheric transmission and laser “brightness” reasons. Overtone efficiencies of 50-70\% relative to the fundamental performance were demonstrated [34-36]. This result is of particular importance because an HF overtone laser with 50\% of the fundamental and having approximately half the wavelength of the fundamental would result in an increase in intensity/brightness due to a $\lambda$ dependence [15].

Graves et al have also reported the results on overtone technology [37]. The main objective was to demonstrate scalability in laser module size through the Overtone Research Advanced Chemical Laser (ORACL): a high power HF Overtone device that can operate in the HF fundamental and DF modes as well. The stress is on the new type of mixing nozzle that improves the laser power density (kW/cm$^2$) and also the laser specific power (kW/kJ reactant) and simplifies manufacturing.

1.3 Literature Survey on Development of Arc Plasma Devices

Thermal plasma is normally generated in equipment called arc plasma generator or arc plasma heater or plasma torch, which consists essentially of two electrodes (cathode and anode). An electric arc is maintained between the electrodes.
A certain amount of gas is injected into the arc plasma heater (in principle any gas can be used) receiving energy from the arc and generating the thermal plasma. An electrical arc plasma heater is inherently capable of heating gases to very high temperature or producing gas plasma, which consists of gas ions with overall neutral charge. It has got ability to produce and handle continuous gas flow at ultra high temperature, which is above 5000 K at high velocity of the order of sonic velocity [38-41].

A specific arc device design and operating conditions are normally needed for the different industrial applications. This normally implies an empirical development of the equipment resulting in high costs and long development period for the construction modification and tests of the specific equipment. A possible alternative for reducing development costs of arc device is to understand the phenomena involved in the arc device operation and using mathematical models and numerical schemes solve the resulting equations. Not only this approach reduces the time spent for developing the arc device but also reduces the direct costs involved in an empirical development. The resulting physical and mathematical models of arc are used for different applications simulating different sets of operating conditions and arc device geometries [42-46].

1.3.1 Brief history of thermal plasma gas discharges

High intensity gas discharges are also called electric arcs, because the first discharge observed in detail was a carbon arc burning in horizontal position in the ambient atmosphere and thus bent by natural convection. The arc discharge not only forms a basis for industrial applications and research activities including laser research, it can also be an initiator for the development of plasma physics itself. From discovery of a certain phenomenon (e.g: the use of metal halides for lighting
purposes) to it's industrial use it can take more than 60 years and up to now, most practically important discharges cannot be computed in advance and thus may not be regarded as fully understood.

JD Cobine and EE Burger published an analysis of the phenomena at the electrodes of a high-current electric arc [47]. In 1956, Finkelburg and Maecker brought out detailed document on theory of electric arc and thermal plasma in German Handbook of Physics [48]. The initial interest in thermal plasma technologies took place in the 1960's and was associated with aerospace programs. Richard R John and William L Bade published paper on “Recent Advances in Electric Arc Plasma Generation Technology” in American Rocket Society journal depicting the engineering use of electric arc for heating gases to high temperatures from the beginning of last century [49]. The 1970's have seen the development of industrial applications in the field of cutting, welding, spraying, transferred arc reclamation, analysis by inductive coupled plasmas, and tentative developments in melting and refining, extractive metallurgy, ultrafine particle synthesis, powder spheroidization and lighting. JJ Lowke presented analytic expressions for arc voltage, electric field, arc radius, plasma velocity and energy balance equations to give approximate properties of free-burning arcs occurring in various industrial applications [50]. The basic studies on free-burning high intensity argon arc by KC Hsu, K Etemadi, E Pfender and others established boundary conditions, i.e. current density in the vicinity of cathode [51]. The efforts in fundamental research to understand the phenomena in thermal plasma conditions in 1980's renewed the interest for applying thermal plasmas in material processing and waste management [52]. In 1990s, many refereed papers appeared in literature on modeling of atmospheric-pressure arcs used in variety of material processes by Maher I Boulos and others [53-60]. Last decade has
witnessed considerable advances in theoretical and experimental investigations of electrode phenomena in high-pressure arc discharges and many review papers got published by various authors [61-64].

Given below is collection of some of the important milestones of recent work/applications of arc plasmas including development of chemical lasers.

<table>
<thead>
<tr>
<th>Year &amp; Contributor</th>
<th>Contribution</th>
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</thead>
<tbody>
<tr>
<td>1970s Spencer DJ</td>
<td>use of arc heaters for development of CW chemical lasers [28]</td>
</tr>
<tr>
<td>1982 Tom Foster &amp; G Lin</td>
<td>plasma arc heater design [65]</td>
</tr>
<tr>
<td>1997-2000 Pierre Fauchais</td>
<td>fundamentals &amp; applications of thermal plasmas [66]</td>
</tr>
<tr>
<td>1998 Andreas Schutze</td>
<td>review of atmospheric-pressure plasma jet [40]</td>
</tr>
<tr>
<td>2001 Ki Chen &amp; H Li</td>
<td>heat transfer and fluid flow in free burning arcs [67]</td>
</tr>
<tr>
<td>2002 Venkatramani</td>
<td>industrial plasma torches and applications [68]</td>
</tr>
<tr>
<td>2004-06 F Lago et al</td>
<td>numerical modeling of an electric arc [69-71]</td>
</tr>
<tr>
<td>2005 A Gleizes et al</td>
<td>review on thermal plasma modeling [46]</td>
</tr>
<tr>
<td>2006 R Bini et al</td>
<td>numerical and experimental study on transferred arcs [72]</td>
</tr>
<tr>
<td>2007 JP Trelles et al</td>
<td>modeling of arc reattachment process in plasma torches [63]</td>
</tr>
<tr>
<td>2008 Benilov</td>
<td>modeling of plasma-electrode interaction in high-pressure arcs [64]</td>
</tr>
<tr>
<td>2009 Murphy et al</td>
<td>review on modeling of thermal plasma for arc welding [62]</td>
</tr>
<tr>
<td>2011 Masaya Shigeta</td>
<td>thermal plasmas for nanofabrication [73]</td>
</tr>
</tbody>
</table>
1.3.2 Brief introduction of thermal plasma gas discharges

The plasma state is frequently referred to as the fourth state of matter in sequence: solid, liquid, gas and plasma. The high energy content of plasma compared to that of solids, liquids, or ordinary gases lends itself to number of important applications. The definition of the plasma state will be restricted to gaseous plasmas, which consists of a mixture of electrons, ions and neutral particles. Since the masses of ions and neutrals are much higher than the electron mass, neutrals and ions are classified as heavy particles or the heavy component in plasma. Particles in an excited state can return to their ordinary or ground state by photon emission. The latter process is at least partially responsible for the luminosity of plasma. In addition to ions and neutral particles, plasma also contains excited species and photons, i.e., in general plasma consists of electrons, ions and neutrals in the ground state, excited species and photons. Such a mixture, however, qualifies as plasma only if the negative and positive charges balance each other, i.e., overall plasma must be electrically neutral. This property is known as quasi-neutrality.

In contrast to an ordinary gas, plasmas are electrically conducting due to the presence of free charge carriers. In fact, plasmas, may reach electrical conductivities exceeding those of metals at room temperature. For example, hydrogen plasma at one atmosphere heated to a temperature of $10^6$ K has approximately the same electrical conductivity as copper at room temperature [75-78].

Kinetic temperatures in plasma, as in many gaseous medium, are defined by the average kinetic energy of a particle (molecule, atom, ion, or electron), i.e. given as:
\[ \frac{1}{2} m \bar{v}^2 = \frac{3}{2} kT \]  

(1.1)

where \( m \) is the mass of the particle, \( \bar{v} \) is its rms or effective velocity, \( k \) is the Boltzman constant, and \( T \) represents the absolute temperature (K). Equation 1.1 implies that the particles follow a Maxwell-Boltzman distribution.

The most common way to generate and maintain plasma is by means of an electric discharge. In such a discharge the high-mobility electrons pick up energy from the applied electric field and then transfer part of this energy to the heavy particles through elastic collisions. But even with an excellent collisional coupling (high collision frequency) between electrons and heavy particles, there will always be a difference between the electron temperature and the temperature of the heavy species in the plasma. The energy transferred from an electron to a heavy particle in a single collision may be expressed by:

\[ \frac{3}{2} k(T_e - T_h) \frac{2m_e}{m_h} \]  

(1.2)

where \( m_e, m_h, T_e \) and \( T_h \) represent the electron mass, mass of heavy species, electron and the heavy particle temperatures, respectively. The energy that an electron acquires from the electric field \( E \) between collisions is given by:

\[ eE \bar{v}_d \bar{t}_e \]  

(1.3)

where \( \bar{v}_d \) is the average drift velocity and \( \bar{t}_e \) the average free flight time between collisions. With \( \bar{t}_e = l_e / \bar{v}_e \) where \( \bar{v}_e = (3kT_e/\pi m_e)^{1/2} \) and \( l_e \) is the mean free path (mfp) of the electrons, it follows that for a steady-state situation.

\[ \frac{T_e - T_h}{T_e} = \frac{3\pi m_h}{2m_e} \left( \frac{eE}{3kT_e} \right)^2 \]  

(1.4)
According to equation 1.4, kinetic equilibrium \((T_e = T_h)\) requires that the energy acquired by the electrons in an electric field between collisions must be very small compared to the average kinetic energy of the electrons. Another interpretation of equation 1.4 considers the fact that

\[
l_e \sim \frac{1}{p} \quad \text{(p is pressure)}
\]

And thus

\[
\frac{T_e - T_h}{T_e} = \frac{\Delta T}{T_e} \sim \left( \frac{E}{p} \right)^2
\]

(1.5)

This relation shows that the parameter \(E/p\) plays a governing role for determining the kinetic equilibrium situation in plasma. For small values of \(E/p\), the electron temperature approaches the heavy particle temperature - this is one of the basic requirements for the existence of Local Thermodynamic equilibrium (LTE) in plasma.

Thermal plasmas, which are classified as “hot” plasmas in the American and European literature and as “low temperature” plasmas (to distinguish them from thermonuclear fusion plasmas) in the Russian literature, are by definition in or close to Local Thermal Equilibrium (LTE). Over the past years, it has become increasingly clear that the existence of LTE in plasma is the exception rather than the rule. Many plasmas that are classified as thermal plasmas do not meet all requirements for LTE, i.e., they are not in complete local thermodynamic equilibrium (CLTE). Because of the small contribution of excited species to the enthalpy of plasma, this type of deviation from CLTE is immaterial for, most engineering applications. For this reason, such plasmas are still treated as thermal plasmas or, more accurately, as plasmas in partial local thermodynamic equilibrium (PLTE).
As mentioned above, the parameter $E/p$ plays a crucial role in attaining kinetic equilibrium. Kinetic equilibrium becomes feasible for small values of $E/p$, i.e., high pressures and/or small values of $E$. Typically, pressures in LTE plasmas exceed 10 kPa (~0.1 atm), also for pressure below 10 kPa, the electron and heavy-particle temperatures separate ($T_e > T_h$).

For thermal plasmas, the magnitude of the electric field is associated with the electrical conductivity ($\sigma_e$) of the plasma through Ohm's law:

$$j = \sigma_e E$$  \hspace{1cm} (1.6)

where $j$ is the current density. For a given current density, the field strength decreases as $\sigma_e$ increases.

The particle number density can be used instead of the pressure in this parameter, according to Dalton's law

$$p = \sum_r n_r kT$$  \hspace{1cm} (1.7)

provided that LTE exists in the plasma. The subscript $r$ stands for electrons, ions, and neutral particles. If the degree of ionization is very small ($\xi \ll 1$) equation 1.7 reduces to:

$$p = nkT$$  \hspace{1cm} (1.8)

where $n$ is the number density of the neutral particles.

In contrast, non-thermal plasmas are non-equilibrium plasmas and frequently classified as "cold" plasmas, because of the low temperature of the heavy species ($T_h \ll T_e$). In comparison to thermal plasmas, non-equilibrium plasma systems are in most cases operated at pressure $< 10$ kPa. According to $E/p$ criterion, substantial deviations from kinetic equilibrium are expected for large value of $E/p$. GE
Georghiou et al published detailed review of the work carried out on the numerical characterization of non-thermal gas discharge plasma [79]. But for application of chemical laser, arc heater operation is at high pressure and thus LTE condition is assumed.

1.3.3 Characteristics of electric arc

The establishment of the electric arc, whether by separation of current carrying contacts or by momentary ionization of the arc gaps, results in the creation of a conducting arc column, which acts as electrical conductor. This current carrying column consists of three distinct regions (fig.1.4) [77], namely:

1) The conduction zone
2) the cathode fall space
3) the anode fall space

![Fig.1.4 Representation of Arc Regions](image)

The conduction zone consists of equal densities of free electron and positive gas ions drifting toward the cathode and anode respectively, comprises the major positions of the electrode gap, and exhibits a comparatively low and constant voltage gradient. Because of the greater mass of the positive ions, nearly all of the current
transport in the conduction columns is by the high-speed electron travel from the cathode to anode.

The cathode and anode fall spaces are the transition region resulting from current transport between solid and gas media are small in extent and are characterized by steep and variable voltage gradients. Furthermore these fall spaces exhibit a reduction in conduction area from that of the conduction zone and this contact is more pronounced at the cathode than anode, the major contraction fall space. At the cathode, the major contraction creates an excess of positive ions accompanied by a rise in voltage across the fall space, coincident with the area in reduction. In a similar fashion, the electrons are deposited on the anode with a resulting net negative charge and final voltage drop in the anode fall space.

1.3.3.1 Voltage-Current Characteristics:

The voltage-current characteristic of the dc low pressure electrical discharge tube shown in fig.1.5 [77], illustrates the three principal divisions of the arc regime: these include the glow-to-arc transition between point H and I, which is triggered by electron emission from the cathode. This emission results from the high heat loads on the cathode, which occur in the high current density regions of the abnormal glow discharge, around the point H on the curve. When the discharge settles down to the point I on the diagram, which is determined by the internal impedance of the dc power supply and its associated circuit, the arc will usually be in non-thermal, low intensity division of the arc regime. This division is characterized by total currents between approximately 1 and 50 A, although non-thermal, low intensity arcs may operate in rare cases outside the limits of this range. This is a region of negative resistance characteristic: i.e. the current-voltage curve has a negative slope in which an increased current results in a decreased voltage. Beyond point J on the diagram, at
approximately 20 to 50 A, the arc moves into a division of the arc regime with a nearly flat or slow rising voltage-current characteristic, which is called thermal or high intensity division of the arc regime, between point J and K in fig. 1.5. The arc discharge is distinguished by glow discharge by much lower voltage at higher current. The transition from the glow to arc is in general non-smooth. The voltage current characteristics of arc are initially falling and then slowly rising.

![Diagram showing Voltage-Current characteristics of the dc discharge](image)

**Fig.1.5 Voltage-Current characteristics of the dc discharge**

1.3.4 Arc plasma composition and properties

The composition, thermodynamic properties and transport coefficients of thermal plasmas strongly vary with temperature. In this section, a brief overview of the important properties of thermal plasmas is given, starting with the plasma composition. For the behavior of thermal arcs a number of thermophysical properties are of importance, which is usually, sub divided into thermodynamic and transport properties and depends directly on the plasma composition [75]. *The composition and arc plasma properties with respect to temperatures are usually either calculated or taken from available literature for modeling work.*
1.3.4.1 Plasma Composition

For the sake of simplicity, the following considerations will be based on thermal plasmas that contain only one type of ion, namely, singly ionized atoms. If such a plasma is generated from a monatomic gas (for example, argon), then only three species compose the plasma—electrons, neutral argon atoms (some of them may be in an excited state), and positive argon ions (again some may be in an excited state).

\[ \text{Ar} \leftrightarrow \text{Ar}^+ + e \]

The plasma composition in this situation is described by a set of equations: the Eggert-Saha equation (1.9), Dalton’s law (1.10), and the condition for quasineutrality (1.11) of the plasma:

\[ \frac{n_e n_i}{n} = \frac{2Q_i}{Q} \left( \frac{2\pi m_i k T}{\hbar^2} \right)^{3/2} \exp \left( -\frac{E_i}{kT} \right) \]  

(1.9)

\[ p = (n_e + n_i + n)kT \]  

(1.10)

\[ n_e = n_i \]  

(1.11)

In the Eggert-Saha equation 1.9, \( n_e \) is the electron number density, while \( n_i \) and \( n \) represent ion and neutral number densities, respectively, regardless of whether the ions and neutrals are in an excited state or in the ground state; \( Q_i \) and \( Q \) are the partition functions of the ions and neutrals, respectively, \( \hbar \) is Planck’s constant and \( E_i \) represents the ionization energy. The partition functions (or sum over all states) are given by

\[ Q_i = \sum_s g_{i,s} \exp \left( -\frac{E_{i,s}}{kT} \right) \]  

(1.12)
\[ Q = \sum \gamma(\gamma, \frac{E_s}{kT}) \]  

(1.13)

Where \( g_{is} \) and \( g_s \) are the statistical weights of the energy levels of the ions and neutrals, respectively, while \( E_{is} \) and \( E_s \) are the corresponding energy levels of the excited states, the equations for the partition functions imply that the populations of the excited states follow a Boltzmann distribution. The Eggert-Saha equation can be derived from thermodynamic principles (minimization of Gibbs free energy), and therefore it can be considered a "mass action law" for the ionization process. For a given pressure, Equations 1.9 – 1.11 permit the calculation of plasma composition as a function of temperature.

If plasma is generated from a molecular gas (for example, nitrogen) then the number of possible species comprising the plasma will be increased due to the presence of molecular species. The chemical processes that may occur in the plasma will include dissociation of molecules into atoms and ionization of some atoms. The formations of molecular ions will be neglected. The dissociation in nitrogen plasma is given by:

\[ N_2 \leftrightarrow N + N \]

This can be described by an equation similar to the Eggert-Saha equation, i.e. the mass action law for the dissociation process. Taking dissociation, ionization, and the presence of additional species into account, the composition of the nitrogen plasma can be calculated. The results for a nitrogen plasma i.e. plasma density vs temperature at \( p=100 \) kPa are given in literature. For \( T >10^4 \) K, nitrogen molecules are no longer present due to dissociation, and ionization of nitrogen atoms reaches a
peak around \( T = 1.5 \times 10^4 \) K. For temperatures \( T > 2 \times 10^4 \) K, the plasma is in practical terms, fully ionized, i.e., the number density of atoms becomes negligible.

1.3.4.2 Thermodynamic Properties

The thermodynamic properties of plasmas include the mass density, the internal energy, the enthalpy, the specific heat and the entropy. In addition, there are derived thermodynamic functions i.e. the Helmholtz function (free energy) and Gibbs function (free enthalpy or chemical potential). The mass density \( \rho \) follows directly from the plasma composition as:

\[
\rho = \sum_n n_i m_i
\]  \hspace{1cm} (1.14)

where \( n_i \) refers to mass number density of various species present in the plasma and \( m_i \) represents the corresponding mass. The other thermodynamic functions, including the derived functions, can be calculated from the partition functions, which play a crucial role in the evaluation of thermodynamic functions. The partition functions establish the link between the coordinates of microscopic systems and macroscopic thermodynamic properties. With the calculated plasma composition, thermodynamic properties of plasma can be calculated (mass density, internal energy, enthalpy, entropy and specific heat) provided that the partition functions for the various species are known.

The partition functions establish the link between the coordinates of microscopic systems and macroscopic thermodynamic properties. In general, the partition function of a particle can be expressed as

\[
Q = \sum_S g_s \exp\left(-E_s/kT\right)
\]  \hspace{1cm} (1.15)
Where $E_S$ represents all forms of energy that a particle can assume and $g_s$ accounts for the degeneracy or statistical weight of each energy level.

It is customary to divide the energy of a particle into translational energy ($E_{str}$) and internal energy ($E_{int}$), i.e.

$$E_S = E_{str} + E_{int} \quad (1.16)$$

These energies are associated with the corresponding translational and internal degrees of freedom of a molecule. The latter include electronic excitation, rotation, vibration, nuclear spins, and chemical reactions. In the Born-Oppenheimer approximation that is valid for gases and plasmas, the total internal energy of a molecule can be expressed as the sum of all the previously mentioned energies. Thus the total partition function $Q$ of a molecule can be expressed by a simple product

$$Q_S = Q_{str} \cdot Q_{rot} \cdot Q_{vib} \cdot Q_{el} \cdot Q_{nuc} \cdot Q_{ch} \quad (1.17)$$

where the individual partition functions represent translational, rotational, vibrational, electronic, nuclear and chemical contributions, respectively.

The translational partition function can be obtained by integration over all spatial and momentum coordinates of a molecule give

$$Q_{str} = \frac{V}{h^3} (2\pi mkT)^{3/2} \quad (1.18)$$

where $V$ is the volume of the system and $m$ the mass of the molecule.

**1.3.4.3 Transport Properties**

Transport phenomena in plasmas encompass the flow situation of every plasma constituent, namely electrons, ions and neutrals, including radiation fluxes under the influence of driving "forces" as for example, electric fields, temperature, pressure,
density and velocity gradients. In order to describe the transfer of electrical charge, mass, momentum and energy within a plasma and from a plasma to its surroundings, characteristic transport properties have been defined as for example, electrical conductivity, heat conductivity, viscosity, diffusivity and optical emission coefficient. Many theoretical considerations in plasma physics are based on the assumption of uniform plasmas. It is, however, very difficult—if not impossible—to produce such uniform plasmas. As mentioned above actual plasmas will reveal gradients in such characteristics as particle number densities \((n)\), applied electrical potentials \((V)\), Temperature \((T)\), and velocity components \((v_x)\). These gradients can be considered “driving forces” that give rise to fluxes. If the magnitude of these gradients remains within certain limits, there will be linear relationships between the driving forces and the fluxes. Examples of such relationships are:

\[
\vec{\Gamma} = -D \text{ grad } n \quad (1.19)
\]

Ohm’s law
\[
\vec{j} = -\sigma \text{ grad } V \quad (1.20)
\]

Fourier’s law
\[
\vec{q} = -k \text{ grad } T \quad (1.21)
\]

And
\[
\vec{f}_x = -\mu \text{ grad } v_x \quad (1.22)
\]

Where \(\vec{\Gamma}, \vec{j}\) and \(\vec{q}\) refer to fluxes due to diffusion, electrical conduction, and thermal conduction, respectively. The term \(\vec{f}_x\) represents the frictional force in the x direction. These linear relationships between fluxes and driving forces incorporate the so-called transport coefficients \(D, \sigma, k\) and \(\mu\), which are known as the diffusion coefficient, the electrical conductivity, the thermal conductivity, and viscosity, respectively.

Last few decades have also shown tremendous progress in the field of laser technology. Another important application of arc plasma devices was established in
1970's for the development of gas dynamic and chemical lasers. The chemically pumped lasing action from arc driven HF laser system was demonstrated by Spencer et al., for which they were granted US patent [28]. As discussed above, the authors published numerous papers for performance and optimization of lasing parameters with Nitrogen or Helium diluents [80-82]. Also during this time at French-German Research Institute of Saint-Louis (France), Andre Sontag et al. [19, 83] reported construction and operation characteristic of an arc-driven supersonic CW chemical laser for kW level laser power. In one of the recent work published as master’s thesis of National Defense University of Science & Technology, China has reported study on arc-driven fluorine atom generator for chemical laser utilizing low power (3 kW) arc heater for optimization of lasing parameters [84]. These researchers utilized the 3 to 140 kW arc heaters for different laser powers. However, they have not given details on development of arc plasma devices. Also there is no open literature available on development of arc plasma devices for chemical laser application.

The arc plasma devices are effectively used for study of various parameters responsible for lasing action in gas/chemical lasers.

1.4 Aim and Motivation of Research Work

For development of arc-driven HF/DF Laser, the basic principle of operation consists first in generating F atoms in a gas mixture through arc plasma, then cooling the F atoms gas mixture through supersonic nozzle and then mixing Hydrogen/Deuterium thus yielding vibrationally excited HF/DF. The Sulfur Hexafluoride (SF₆) is chosen as fluorine carrier because it is non-toxic and non-corrosive gas and can be handled safely in large quantities. For development of a kW level HF/DF laser, a total mass flow rate of 5-7 g/s of gas mixture (Nitrogen, Sulfur Hexafluoride and Oxygen etc) is desirable from the first principle of chemical reaction.
A 50 kW arc plasma heater is sufficient for 5-7 g/s of gas mixture flow rate to attain desired temperature and pressure suitable for a kW level chemical laser. The present work aims at the design & development of 50 kW arc plasma heater suitable for a kW level HF/DF laser. While designing arc heater for chemical laser applications, following basic requirements have to be considered:

i) The design and selection of electrodes (cathode and anode) for minimum erosion (to attain pure chemical composition of lasing gas mixture)

ii) Rotation of the arc for distribution of anode heat load and thereby reducing the anode erosion

iii) Uniform heating of large volume of gases (5-7 g/s) at temperatures of about 2000 k

iv) The stable arc operation for 10-20 seconds

Arc devices having the topology with following features meet these requirements:

a) Approximate rotational symmetry about a central axis

b) Co-axial electrodes separated by an angular gap across which the arc current passes, and through which the gas flows

1.4.1 Motivation for Present Research Work

The most widely used arc plasma devices are plasma torches used for number of industrial applications such as welding, cutting, spraying and waste treatment etc [78]. There are different type and geometries of plasma torches. The two main classes of plasma torches are so called transferred and non-transferred torches. In the transferred arc, the plasma is maintained between one electrode of the torch (normally the cathode) and a piece of metal (or another conducting material) that is to be cut or melted located outside the torch. Whereas, non-transferred plasma torches employ the two electrodes of the torch in order to maintain the electric arc, i.e. the electric arc
strikes between the two electrodes of the torch and it is kept inside the torch. These plasma torches are normally used for spraying, production of advanced materials and treatment of hazardous waste etc. Typical power levels of plasma torches used for spraying are between 10 and 100 kW. Argon is normally employed as plasma gas since it is an easily ionized and it does not react with the material being treated or sprayed. In general, the plasma torches have been designed for different applications for power levels from a few kW to approximately MW levels. Thermal efficiencies for different operating conditions are in the range from 30 to 90%. The maximum temperature in the plasma jet is a function of the operating parameters and may vary from 8000 to 20,000 k close to nozzle orifice. Since the plasma jet in this type of torches is field-free plasma, the plasma temperature decays rapidly with increasing distance from the nozzle orifice. These geometries of plasma torches are very well utilized for various industrial applications. However, these plasma torches do not meet the requirements of large flow rates and pressure required for laser applications.

Although researchers have used arc plasma devices for initial performance of CW chemical lasers, but technical details on the development of arc plasma devices for laser applications is not available in the open literature. Moreover, control, sensors and data acquisition aspects for hostile arc-driven HF/DF chemical laser are not addressed anywhere as most of these systems have been developed for defense applications. Also chemical reaction in arc-driven HF/DF laser produces highly toxic gases like HF and atomic fluorine, therefore their handling is a tough task in general and needs more attention when it comes to lab environment. Any human error may prove fatal and hence automated safety and interlocks with sufficient redundancy should be incorporated with DAS for prompt action. HF/DF chemical laser system is a complex system involving many important sub systems such as arc heater system,
gas supply system, cooling system, power supply system, vacuum and pressure recovery systems etc. All these sub systems may be located at wide spread isolated locations in dedicated rooms. Therefore, for the ease of operation single person control console is essential for minimizing the risk factor. This in turn requires all monitoring parameters and control on the intelligently managed window screen.

1.4.2 Methodology

For chemical laser applications, the non-transferred arc plasma torch has to be modified to cater large volume of gases and to have geometry for uniform heating of arc plasma gases with less electrodes erosion. The configuration of arc heater for chemical laser should be concentric cylindrical type electrode configuration (with free arc length design) consisting of a thoriated tungsten water-cooled central cathode and a surrounding water-cooled Oxygen Free High Conductivity Copper (OFHC) anode. A magnetic field coil may be incorporated on the anode for distribution of the anode heat load over large area. In this way the lifetime of the anode is increased and the level of contamination of the plasma is reduced. This configuration of arc plasma heater is most preferred geometry for chemical laser application and will be meeting all the basic requirements. In order to further reduce electrode wear, electrodes may be over-protected by efficient cooling systems. This measure, however, reduces the efficiency to some extent of the arc plasma heater, especially for the long duration continuous operations. But for arc operation for short durations of the order of tens of seconds, as is the situation in the present case, efficient cooling at the cost of slightly low arc plasma heater efficiency is acceptable for continuous wave HF/DF chemical laser application.

Since the hardware development is both time consuming as well as involves high costs, a possible alternative is to understand first the phenomenon involved in arc
operation using available mathematical models and numerical schemes applicable to arc plasma heater for HF/DF laser application. The governing partial differential equations for mass, momentum, energy and current can be solved for the present geometry with commercially available code COMSOL. The four equations determine the principal arc quantities of temperature, plasma velocity and electric potential for the given arc current. The material functions of the plasma namely thermal and electrical conductivity, viscosity, density and specific heat have to be incorporated as a function of temperature. With the availability of modern high-speed computers, and the careful use of recently developed numerical methods, this level of detailed computation is quite feasible.

As the geometry of each designed arc plasma tunnel is governed by specific application considerations and once the applicable computational model is validated, it can be employed for optimizing the parameters of the tunnel suitable for the given application.

1.4.3 Work Carried Out/ Organization of Thesis

- For performance studies and optimization of lasing parameters on a kW HF/DF arc-tunnel, following developmental work is essentially needed:
  - The modification of arc plasma devices to suit laser applications
  - Development of 50 kW arc heater based on modifications and its validation using modeling studies.
  - Development of laser tunnel that includes plenum, nozzle, optical cavity, diffuser, evacuation and scrubber system etc.
  - Development of dedicated Data Acquisition system for remote operation of arc heater and laser tunnel.
• Interfacing of Arc heater and laser tunnel for detailed performance studies for its suitability for laser applications.

The development and performance studies of this new 50 kW arc tunnel are the major aims of this work presented here. This Thesis has been divided into six chapters:

Chapter 1 gives a brief introduction about Lasers in general and HF/DF chemical lasers in particular. This chapter covers various development stages of arc plasma devices in details along with review of arc-driven HF/DF chemical lasers. This chapter also discusses about the basic arc characteristics and arc plasma properties. The history of arc plasma devices and their industrial applications are also presented along with applications of arc plasma devices to lasers. Finally, the motivation and aim of this present research work has been elaborated.

Chapter 2 discusses mainly the design philosophy and method adopted for realization of arc plasma generator or arc plasma heater for a kW level HF/DF chemical laser. Further, mathematical modeling studies of arc plasma heater geometry suitable for chemical laser applications with commercially available software COMSOL has been carried out. Numerical modeling results of arc discharge phenomenon relevant for HF/DF chemical laser are discussed in details to emphasize the importance of computational work. The comparison of simulation results with studies available in the literature has been made to validate the design adopted in the present studies.

Chapter 3 describes the hardware development 50 kW arc tunnel in details along with diagnostic system. The development of various sub systems of 50 kW arc heater and selection of electrodes materials for less contamination are elaborated in this chapter. Also the development of components such as supersonic nozzle, laser cavity, diffuser and vacuum dump etc required for ultimate interfacing of arc heater with laser tunnel are also discussed in this chapter.
Chapter 4 discusses about the design philosophy and scheme adopted for realization of present dedicated Data Acquisition System (DAS) for noisy and hostile arc environment for arc-driven HF/DF chemical laser. The important requirements from the point of view of operation of HF/DF chemical laser were identified and the corresponding DAS features are also covered in this chapter. Further, different building blocks of PC based DAS are discussed in details along with diagnostic system used for parametric studies on arc tunnel. The importance of various sub systems i.e. gas feed system, cooling system, power system and arc heater system from the DAS point of view is also described in this chapter.

Chapter 5 discusses the integration of all sub-systems of 50 kW arc tunnel for conducting lasing experiments. This chapter also presents the results such as efficiency as function of arc input power, V-I characteristic of arc, effect of mass flow rates of H\textsubscript{2} & SF\textsubscript{6} on arc power and temporal variation of Mach number etc obtained from parametric studies on 50 kW arc tunnel. The performance of 50 kW arc plasma heater has also been evaluated as a function of buffer gas like Argon and Nitrogen in this chapter. The experimental results are discussed in detail and the performance of arc plasma devices for development of HF/DF chemical laser is analyzed.

Chapter 6 presents the conclusion of this research work and future scope of research work in this area. 50 kW arc plasma heater has been designed and developed for a kW level HF/DF chemical laser application. A dedicated custom-built Data Acquisition System (DAS) has been developed and interfaced with arc-driven HF/DF laser tunnel.