國 立 交 通 大 學

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碩 士 論 文

Experimental Study of UV Emission of a Vacuum Ultra Violet Excimer Lamp

以實驗的方法研究真空紫外光準分子燈管之紫外光放射

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指導教授：吳宗信　博士

中華民國九十六年七月

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陳又寧　謹誌

九六年七月於風城

以實驗的方法研究真空紫外光準分子燈管之紫外光放射

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摘　要

介電質放電 (Dielectric Barrier Discharge, DBD)已經發展了很長一段時間了，在許多方面都有廣泛的應用。其中，準分子燈的應用在最近幾年更是受到相當大的注意，由於其在工業上廣泛的應用。像是材料沉積技術、光化學技術、螢光燈管及電漿顯示器等方面。

為了應付越來越廣泛的應用，提升光強度成為一個追求的目標。在本實驗中，我們利用純氙氣及惰性氣體混合來探討混合氣體比例對於光強度的影響。並且調整氣體總壓、頻率及輸入功率等參數，來觀察光強度的變化。

最後，了解各個參數與光強度之間的關係，並調整參數以獲得最佳的光強度表現。

Experimental Study of UV Emission of a Vacuum Ultra Violet Excimer Lamp

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Abstract

Dielectric barrier discharge (DBD) has developed for a long time, and has widely application in many fields. Among them, the excimer lamp attracts goodliness attention in recent years due to its widely application in industry, for example, new materials processing, photochemistry, fluorescent lamp and plasma display panels.

In order to deal with more and more application, to promote light intensity of 172 nm becomes a questing goal. In this experiment, we discuss the effect of mixture fraction on light intensity by means of put in pure Xe and rare gas mixture. And to adjust gas total pressure, frequency and input power to observe the change of light

intensity.

Finally, we can realize the relation between the factors and the light intensity, and obtain the optimized operating conditions.

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Chapter 1 Introduction

1.1 Motivation

Energetic UV photons can initiate a variety of chemical, physical or biological processes. Some examples are photochemical synthesis and degradation, the UV initiated polymerization of coatings and paints, the UV induced deposition of metals and dielectrics and the disinfection of drinking water[Xu, 2001]. A high UV light intensity is required to apply energetic UV photons to industrial applications. The UV spectrum of the source usually has to match the intended process. Intense UV radiation is commercially available from medium and high pressure arc discharge in rare gas and mercury / rare gas mixtures. Selective UV radiation of certain wavelengths is required for many desired processes. In the present thesis, we are interested in measuring the characteristics of an excimer VUV lamp, which is more environmentally friendly as compared to previous mercury lamps.

1.2 Background

1.2.1 Introduction to DBD

Dielectric barrier discharges, or silent discharge, have been known for more than a century. First experimental investigations were reported by Siemens in 1857, see figure1.1. They concentrated on the generation of ozone. This was achieved by subjecting a flow of oxygen or air to the influence of a dielectric barrier discharge (DBD) maintained in a narrow annular gap between two coaxial glass tubes by an alternating electric field of sufficient amplitude. The novel feature of this discharge apparatus was, that the electrodes were positioned outside the discharge chamber and were not in contact with the plasma. In his later years Werner von Siemens considered his discharge configuration for the generation of ozone as one of his most important inventions.

Dielectric barrier discharge (DBD) or silent discharge is a typical non-equilibrium high pressure ac gas discharge, see figure1.2. It is known that the DBD can be occurred between two electrodes, at least one of which should be covered with dielectric, when an ac high voltage is applied on the electrodes as figure1.3. Dielectric barrier discharge (DBD) are coupled high pressure discharges operated from an a.c. high voltage supply at frequencies from 60Hz up to about 1MHz. With increasing voltage, the reaction gas of the system will breakdown, and product many streamers between the electrodes. We can observe luminous phenomenon at the same time. Due to the high pressure of 0.1 to beyond 1 bar the discharge consists of narrow transient plasma microstreamers, which are often distributed statistically both in time and within the space between the electrodes. The microstreamers have diameters of the order of 100μm and usually very short-lived, on the order of 100 ns or loss. Both the exact plasma wire diameter and precise time duration depend upon the gas used and the reactor pressure. In DBD, the dielectric surfaces serve the role of a capacitor in series with the plasma. The plasma in DBD consists of a lot of microstreamers, see figure1.4. When the microstreamers cross the gap and impinge on the dielectric, the dielectric charges up. Since the transverse mobility of charge on the dielectric is extremely low, the charging of the dielectric is restricted to the local vicinity of the streamer. When the local dielectric charges and reduces the voltage across the gap, the streamer is quenched, thereby preventing formation of an arc. The discharge gap itself has a typical width ranging from less than 0.1mm to several centimeters, depending on the application. To initiate a discharge in such a discharge gap filled with a gas at about atmospheric pressure, voltages in the range of a few hundred V to several kV are required. The gas can either flow through the DBD (ozone generation, surface treatment, pollution control) or it can be recycled (CO_2 lasers) or fully encapsulated (excimer lamps, excimer based fluorescent lamps and light panels, plasma display panels).

1.2.2 Introduction to rare gas excimer lamp

The recent use of silent discharges to generate narrow band UV radiation based on excimer formation in the plasma provides a new optical source or lamp for investigating photo-physical and photochemical processes involving the interaction of

UV radiation with matter. Many UV systems based on this technology are used for germicide treatment of surfaces, surface modification, UV curing, and material deposition processes, as well as for UV-induced chemical synthesis and decomposition. For industrial applications excimer UV sources driven by silent discharges may have definite advantages, of high reliability, scalability to large area and very high UV powers, and reduced costs per UV photon. Table 1.1 lists the molecular species which emit VUV and various UV wavelengths.

Excited dimers or "excimers" of argon, krypton and xenon emit narrow-band VUV radiation between 100 and 200 nm. These pure rare gas excimers are efficient fluorescers, converting the electron energy into VUV radiation. Molecular xenon has a slightly bound excimer state at about 8 eV, as shown in figure 1.5. Into this bound state, a large population of states converges via bi-atomic collisions:

$$Xe^{*} + Xe \rightarrow Xe_2^{*} \quad (1.1)$$

The spontaneous emission of photons resulting from a transition from this level to the ground state peaks provides photons around 7.2 eV or 172 nm. In the microdischarges the electron energy has to be optimized for efficient excitation of the atomic Xe^{*} level which can react with neutral Xe atoms to form the Xe_2^{*} excimer.

$$e + Xe \rightarrow Xe^{*} + e \quad (1.2)$$

$$e + Xe \rightarrow Xe^{**} + e \quad (1.3)$$

$$e + Xe \rightarrow Xe^{+} + 2e \qquad (1.4)$$

$$Xe^{*} + Xe + Xe \rightarrow Xe_2^{*} + Xe \qquad (1.5)$$

$$Xe_2^{*} \rightarrow Xe + Xe + h\nu(7.2eV) \qquad (1.6)$$

Since excimer formation is usually a three body reaction, higher pressures favor excimer formation with corresponding decrease in Xe^{*} densities. For this reason non-equilibrium discharges at pressures above 50 torr are required. In addition to the direct path of reactions that form Xe_2^{*} upper states (1.2) and (1.5). We also have Xe_2^{*} state formation by all reactions starting from higher lying excited and ionic atomic and molecular states. Extended kinetic models treating the interaction of high-energy electron beams with xenon are based on the assumption that the various excited states of the xenon atom and molecule can be represented by two (fictitious) excited atomic and molecular states: Xe^{*}, Xe^{**}, Xe_2^{*}, Xe_2^{**}. Silent discharges in xenon can be sources of VUV radiation peaking at 172 nm, having a half-width of 12–14 nm and emitting practically no other radiation in the wavelength region between 180 and 800 nm. Thus, for the purposes of photochemistry, this is a VUV source of high spectral purity. Silent discharge configurations are quite flexible with respect to the geometrical shape and the electrode configuration. See figure 1.6. Figure 1.6(a) is a planar lamp geometry that could be meters in extent. Figure 1.6(b) is an open co-axial geometry for irradiating rods or cylinders placed within the open cylinder with UV light emitted by the outer annulus. Figure 1.6(c) could be used to irradiate the inner surfaces of cylinders meters long, provided we insert the smaller diameter lamp into the larger diameter cylinder. Figure 1.6 (b) shows this geometry, which has a great potential for irradiating fiber surfaces or gas and liquid flows. Hence, a major advantage of the silent discharge is its flexibility to achieve lamps radiating in unique geometric directions with respect to the geometrical shape of surface to be irradiated. As Figure 1.6 shows we can adapt the form of the UV source to the intended surface. In addition to forming plane panels for the irradiation of large surfaces and cylindrical sources radiating outwards or even radiating inwards can be built.

1.3 Literature survey

There have been considerable demands for the development of short incoherent light source in vacuum ultraviolet (VUV), that could produce substantial output with reasonable operation efficiency in compact sizes. Such VUV light source would be applicable to various scientific fields, such as new types of materials processing[Boyd, et al. 1997], photochemistry[Hozumi, et al. 2000], fluorescent lamp[Jinno, et al. 2003] and plasma display panels[Bu, et al. 2002]. Currently available VUV light sources such as deuterium and mercury lamps limit their spectrum and temporal emission characteristics, leading to very limited application fields of these lamps. We can obtain

UV/VUV source by means of gas discharge of rare gas[Kogelschatz, et al. 1990] or rare gas/halogen[Zhang, et al. 1996] mixture. Rare gas excimers have been known to be one of the most efficient VUV source. High tunable laser operation has been demonstrated[Lam, et al. 2002].

1.4 Specific Objectives of the Thesis

An excimer UV lamp has found numerous applications in industry as reviewed earlier. Its fundamental understanding is thus very important. The specific objectives of the present thesis can be summarized as follows:

1. To measure the 172nm UV emission and electrical characteristics from an excimer lamp with pure Xe;
2. To measure the 172 UV emission and electrical characteristics from a excimer lamp with Xe/He mixture;
3. To discuss the measurements obtained in the above and recommend the optimized operating conditions.

Chapter 2 Experimental method

2.1 Experimental Apparatus

2.1.1 Excimer VUV lamp

The lamp in this experiment is made by the quartz, is a coaxial quartz tube. See figure 2.1. Outer tube radius is 40mm, inner tube radius is 16mm. The gap between outer and inner tube is approximately 12 mm. The gap space between the outer and inner tube is filled with Xe or mixture gaseous species by a gas supply system. The gap space is the region of gas discharging for emmition of 172nm UV light. Inside and outside electrodes are made by stainless steel sheet, which connect to a high voltage cyclic electric power supply system.

2.1.2 Gas supply system

This experiment mainly treat the influence of gaseous mixture on light intensity, therefore the gas mixture system is one of the main equipment. Please see figure 2.2. Before igniting the plasma, pump the pipe and lamp by using the mechanical and turbo pump. Put in the gas when the pressure achieves 10^{-6} torr. The gas mixture system may mix three kind of different gas at the same time at the most, this experiment takes Xe as the base gas, and mixes He. We then discuss on the light intensity change under pure Xe and gas mixture with different fraction.

2.1.3 Power supply system

A cyclic pulse electric power was used to ignite gas discharging in the excimer UV lamp. We altered a pulse width adjustable knob on the power supply system (Fig. 2.3) to change output electric current and in turn to change the output power from the power supply system. The output power is correlated to the input power to the lamp. The adjustable range of the pulse width is from 1.5 μs to 17.5 μs. Increasing the pulsed width will result in increasing the output electric current and power increase, and subsequently increasing the input power to the lamp. In the power supply system, the pulse frequency is also adjustable, and its range is from 20 to 60 kHz. Hence, we can measure the light intensity change by means of adjust different power and frequency. The overall experiment equipment that combines the gas supply system and the power supply system are schematically shown in figure 2.4.

2.2 Experimental Instrumentation

2.2.1 UV meter

The UV light intensity of 172 nm is measured by an UV meter - USHIO UIT-250, see figure 2.5. The unit of light intensity is mW/cm^2. When experimenting, the distance between meter and lamp is fixed at 2 mm, and we measure the light intensity change under different conditions.

2.2.2 Thermocouple

The temperature recorder uses T type thermocouple to measure temperature change with gas discharge, the measure range from room temperature to 200°C. See figure 2.6.

2.2.3 Oscilloscope

The oscilloscope of this experiment is Tektronix TPS2014, using high voltage probe to measure voltage and current of input the lamp, and calculate the input power. Moreover may record the waveform of voltage and current, see figure 2.7.

Chapter 3 Results and Discussion

3.1 Light Emission Due to Pure Xe Gas

The intensity of the 172 nm VUV light of an excimer dielectric barrier discharge lamp that was filled with pure Xe gas was measured. The light intensity values were found to be strongly influenced by lamp parameters, such as enclosed Xe gas pressure, as well as power supply parameters, such as electric frequency and current of input power. The relationship between the light intensity and these factors will be shown and discussed as followed.

3.1.1 Effect of gas pressure

The appearance of the discharging DBD lamps as well as the intensity of 172 nm VUV light are found dramatically influenced by the pressure of Xe gas. As shown in figure 3.1, at low Xe pressure (<200 torr) the DBD lamps look like in glow discharging, and their streamer discharging appearance were not obviously found. In addition, their light intensity is also very low at low Xe gas pressure. With increasing pressure, the streamers become obvious and the light intensity is better. Figure 3.2 shows the intensity of 172 nm UV light increases with the increasing pressure of pure Xe gas. When the Xe gas pressure increases to about 400 torr a maximum light intensity reaches. When the gas pressure is beyond 400 torr, the light intensity decrease with increasing pressure.

In order to promote the light intensity of 172 nm, it must have more Xe_2^*. When Xe_2^* is de-excited back to ground state, it emit 172 nm VUV simultaneously. It would seem that higher collision frequency at a higher pressure, the higher collision frequency promote the three-body collision process result in dimer emittion.

3.1.2 Effect of pulsed frequency of power supply

In order to realize the influence of frequency change on gas discharge and light intensity in pure Xe, hence we operated experiment under two different frequencies (40&60 kHz). As figure 3.3, the higher light intensity can be obtained and input more power under higher frequency. Therefore the higher frequency is helpful to the gas discharge and light intensity performance.

3.1.3 Effect of power input

In order to realize the influence of power on light intensity, we can change power by means of adjusting power current to observe light intensity change. The light intensity increase with increasing power current and absorption power change simultaneously as figure 3.4.

3.2 Light Emission Due to Xe/He Mixture Gas

We take Xe as base gas and mixes with different rare gas to improve the light intensity performance. Mixing He with different ratio and change gas total pressure, frequency and input power to observe the light intensity change. What follows will present the results and discussions about how the light intensity, effected by these factors and about what the difference between mixture gas and pure Xe under different condition.

3.2.1 Effect of gas pressure

About the effect of gas total pressure on light intensity, the discharge situation of the lamp and light intensity are different under different pressure. As shown in figure 3.5, when lower pressure (<200 torr), the streamers appear not obvious and look like glow discharge, the light intensity is lower as well. When the pressure increase, the streamers become obvious and the light intensity is enhanced. As figure3.6, the light intensity approaches saturated when pressure near 400 torr. After 400 torr, the light intensity decrease with increasing pressure. Compare with pure Xe, the light intensity enhanced obviously under the same pressure condition.

3.2.2 Effect of mixture fraction

In order to understand the effect of mixture fraction on light intensity, we take Xe as base gas, and put in He with different fraction to observe the light intensity change. As figure 3.7, the light intensity enhanced obviously when the mixture fraction between 5~10%. When mixture fraction exceed 25%, the light intensity is worse than pure Xe. Compared with pure Xe, the light intensity may improve 10~15% when the mixture fraction is 7%. It would seem that the He has longer lifetime in excited state, causing the collision opportunity to increase with Xe or Xe^*, and produces more Xe^*_2 to enhance the light intensity of 172 nm. The collision process is quite complex, the APPL laboratory is developing the computational code to simulate the collision between molecules, we can simulate the collision process and the light intensity performance precisely in the near future.

3.2.3 Effect of pulsed frequency of power supply

In order to realize the influence of frequency change on gas discharge and light intensity in mixture gas, we operated experiments under two different frequencies (40&60 kHz). As in figure 3.8, the higher light intensity can be obtained and input more power under higher frequency. Therefore the higher frequency is helpful to the gas discharge and light intensity performance.

3.2.4 Effect of power input

In order to realize the influence of power on light intensity, we can change power by means of adjust power current to observe light intensity change. The light intensity increase with increasing power current and absorption power change simultaneously is shown in figure 3.9.

Chapter 4 Conclusions and Recommendation of Future Work

4.1 Summary

As mentioned above, the factors that influence the light intensity of 172 nm is as following: gas total pressure, frequency, input power and mixture ratio and so on.

1. In the gas total pressure aspect, the light intensity increase with increasing total pressure and achieve saturation when pressure is near 400 torr.
2. In the mixture ratio aspect, the discharge situation can be improved after putting in a little He (approximately 7%), and enhance the light intensity obviously up to 10~15%, the maximum light intensity is about 68 mW/cm. When the mixture ratio exceeds 25%, the light intensity begin decreasing with increasing mixture ratio.
3. In the frequency aspect, we find that higher frequency may enhance the maximum of input power and obtain higher light intensity from experimental result.
4. In the input power aspect, basically, the light intensity increase with increasing input power current. The lamp can input more power with a higher gas pressure total.

4.2 Recommendation of Future Work

Based on this study, future work is suggested as following:

1. To put in different gas to observe the light intensity change.
2. To change the gap of excimer lamp to find the effect of the lamp's gap on the light intensity.
3. To set up the equivalent circuit of excimer lamp to understand the electrical effect.

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Excimer	Wavelength/nm	UV Range
Ar_2^*	126	VUV
Kr_2^*	146	VUV
F_2^*	158	VUV
$ArBr^*$	165	VUV
Xe_2^*	172	VUV
$ArCl^*$	175	VUV
KrI^*	190	VUV
ArF^*	193	VUV
$KrBr^*$	207	UV-C
$KrCl^*$	222	UV-C
KrF^*	249	UV-C
XeI^*	253	UV-C
Cl_2^*	259	UV-C
$XeBr^*$	283	UV-B
Br_2^*	289	UV-B
$XeCl^*$	308	UV-B

Table1.1 Peaks wavelengths of different DBD excited excimers.

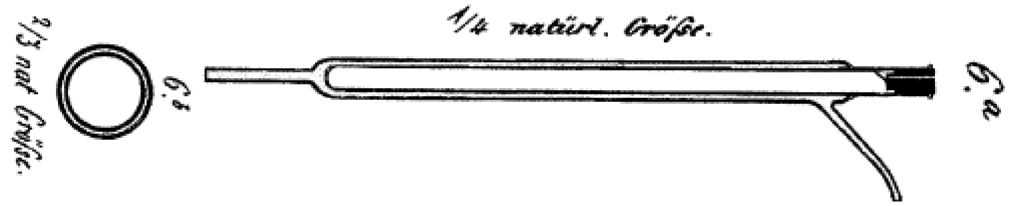


Figure 1.1 Historic ozone discharge tube of W. Siemens, 1857

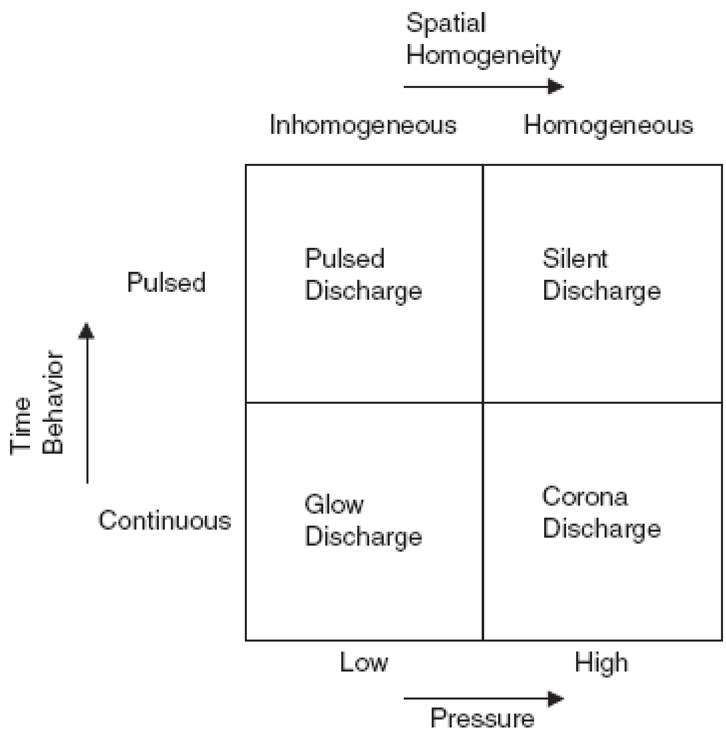


Figure 1.2 Discharge types arranged according to temporal behavior and spatial appearance.

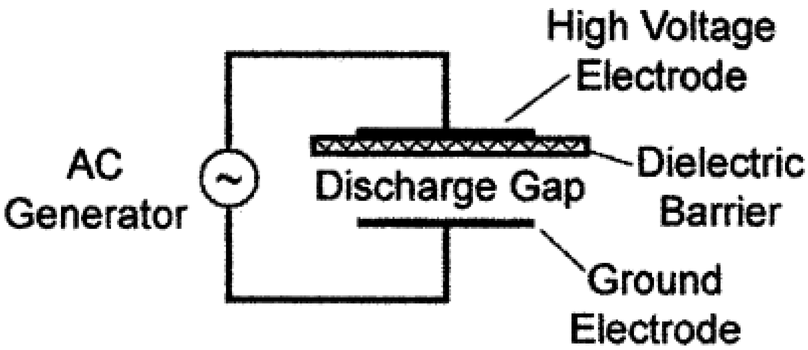


Figure 1.3 Basic dielectric-barrier discharge configurations.

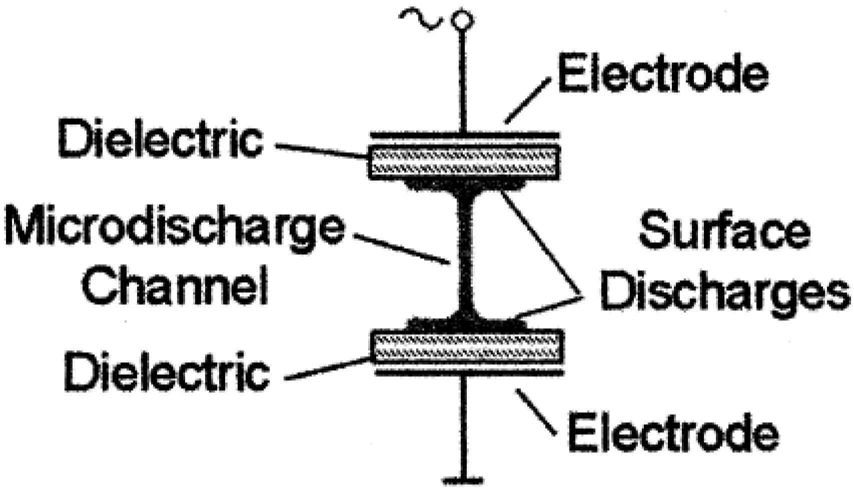


Figure 1.4 Sketch of a microdischarge

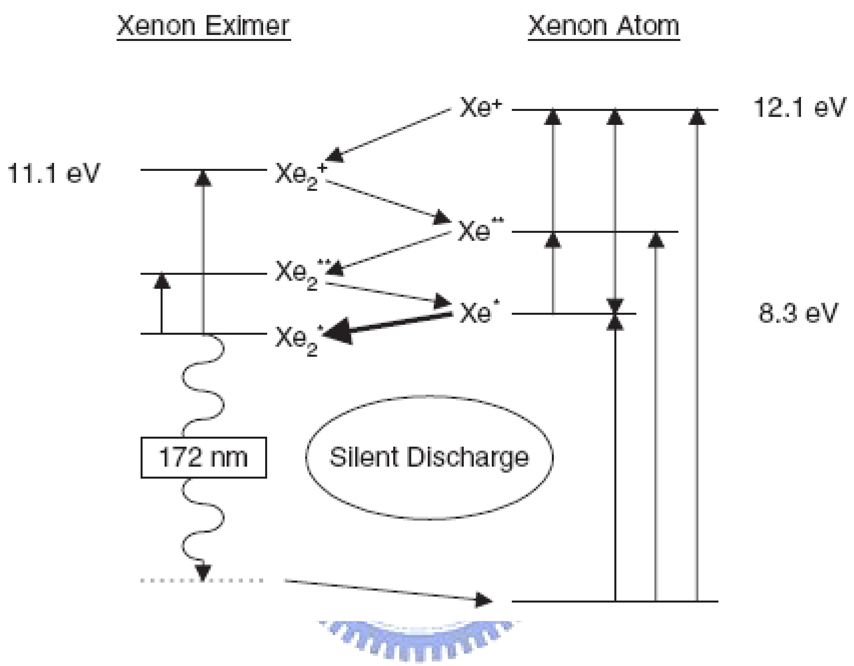


Figure 1.5 Simplified potential energy diagram of Xenon.

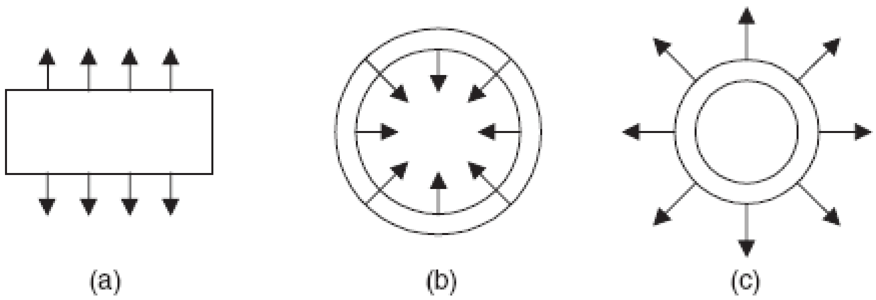


Figure 1.6 Different geometries for silent discharge excimer sources.

Figure 2.1 Excimer VUV lamp.

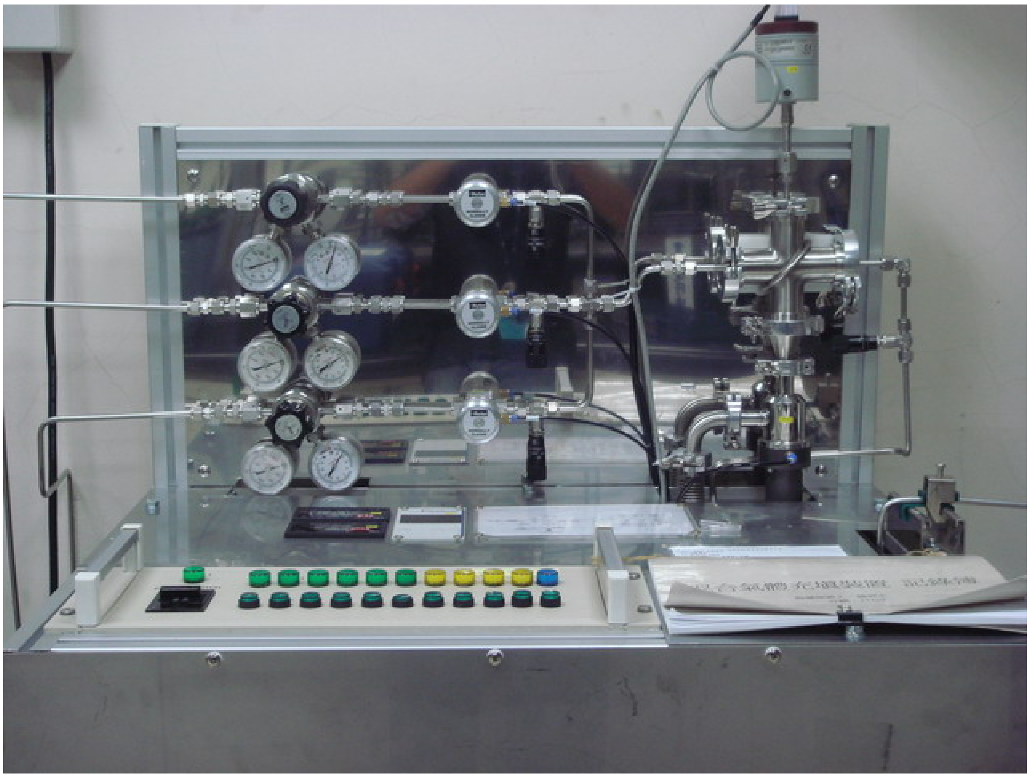


Figure2.2 The gas supply system.

Figure2.3 The power supply system.

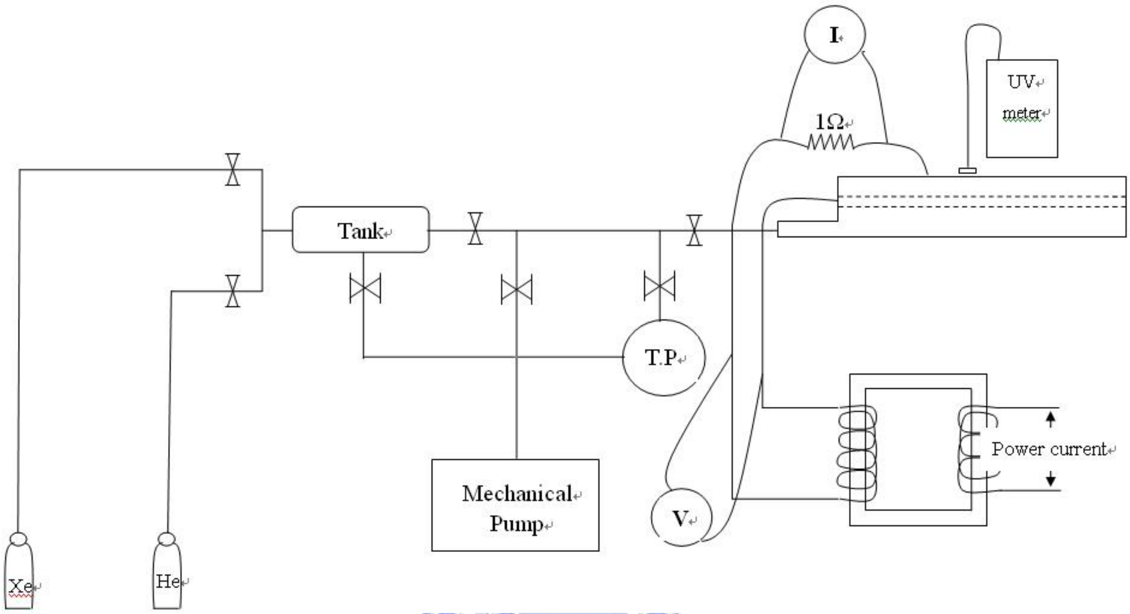


Figure 2.4 Schematic of experimental system.

Figure2.5 The UV meter

Figure2.6 The thermocouple.

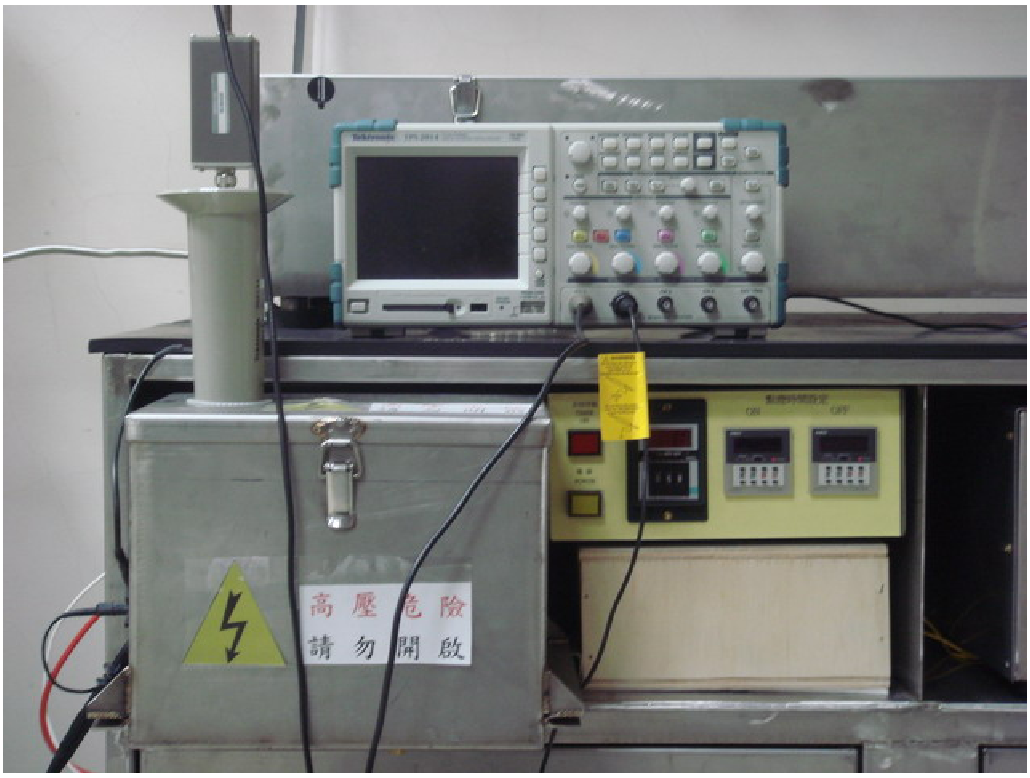
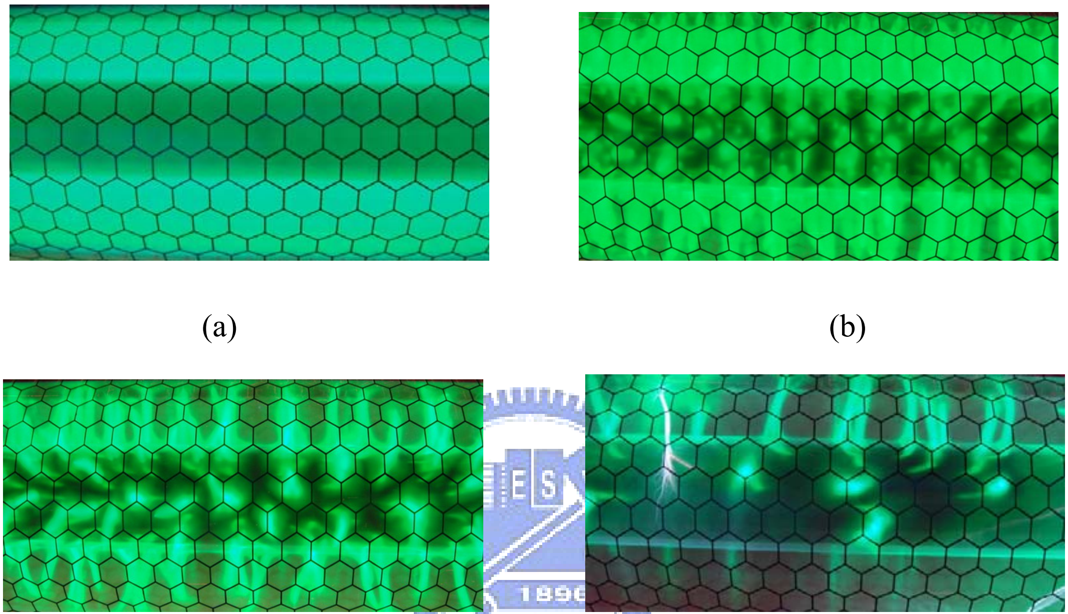


Figure2.7 The oscilloscope and high voltage probe.



(a) (b) (c) (d)

Figure 3.1 The image of discharge situation with different pressure in pure Xe. The pressure is (a) 100, (b) 300, (c) 400 and (d) 500 torr.

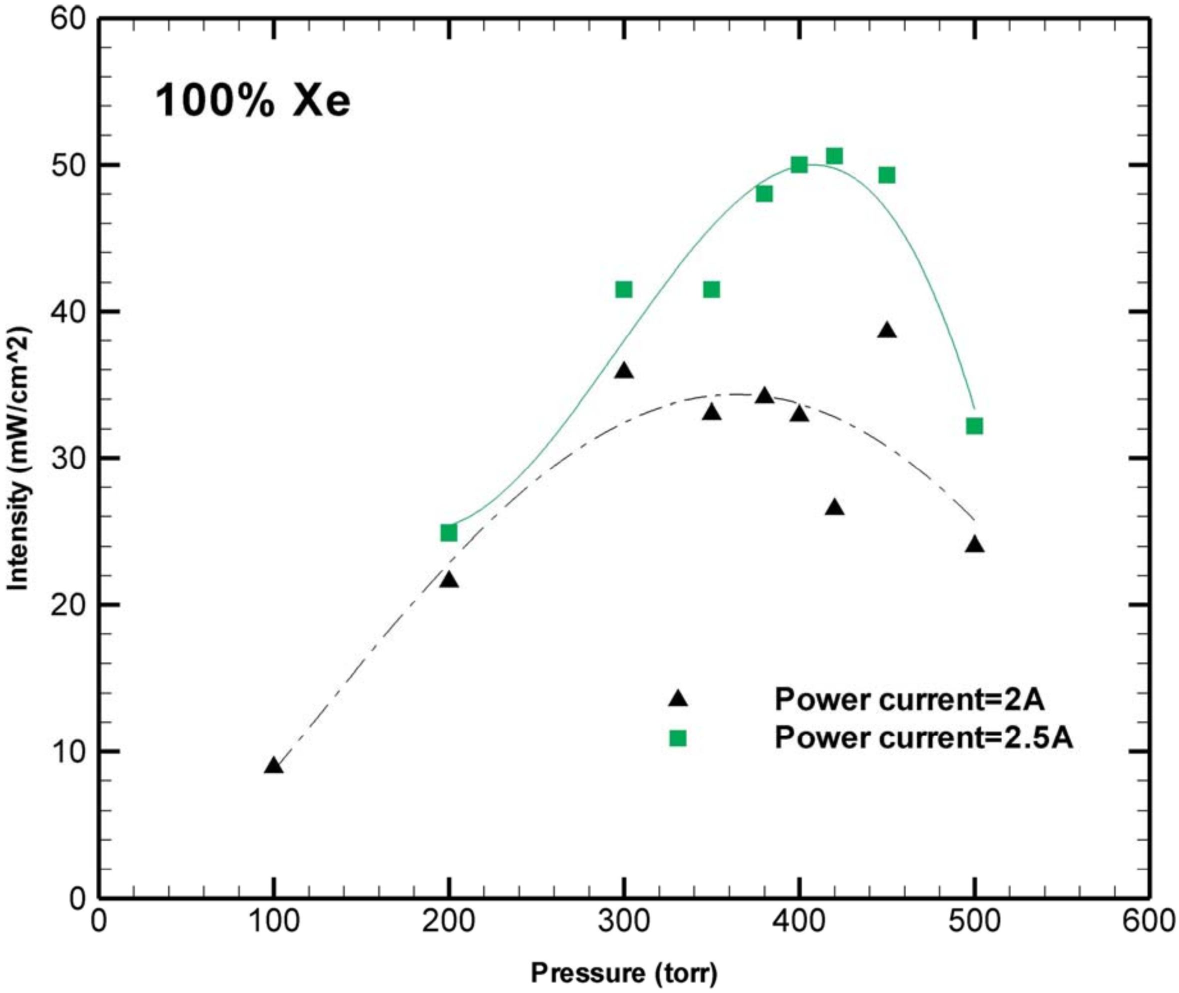


Figure 3.2 The light intensity change with different pressure in pure Xe.

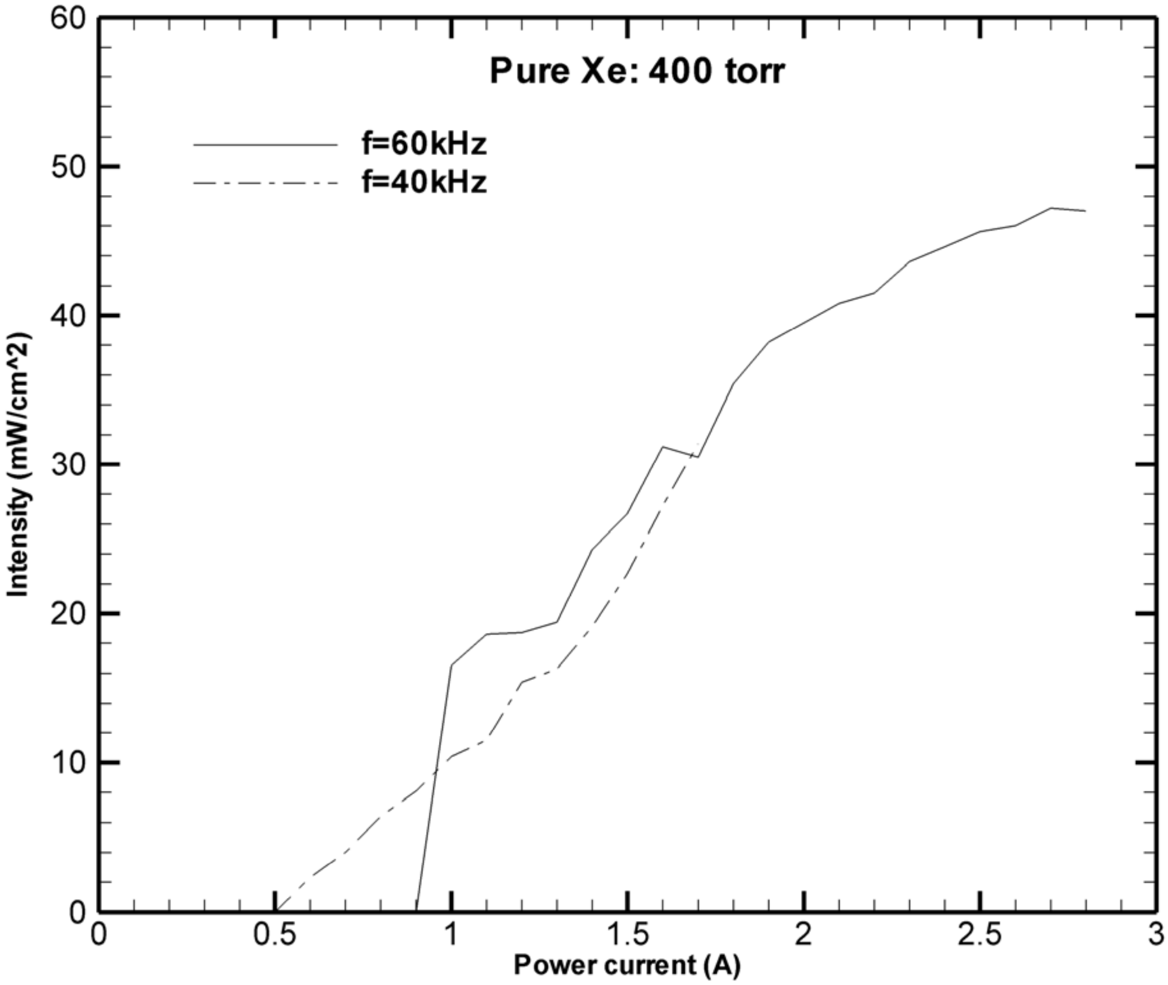


Figure 3.3 The light intensity change with different frequency in pure Xe and pressure

is 400 torr.

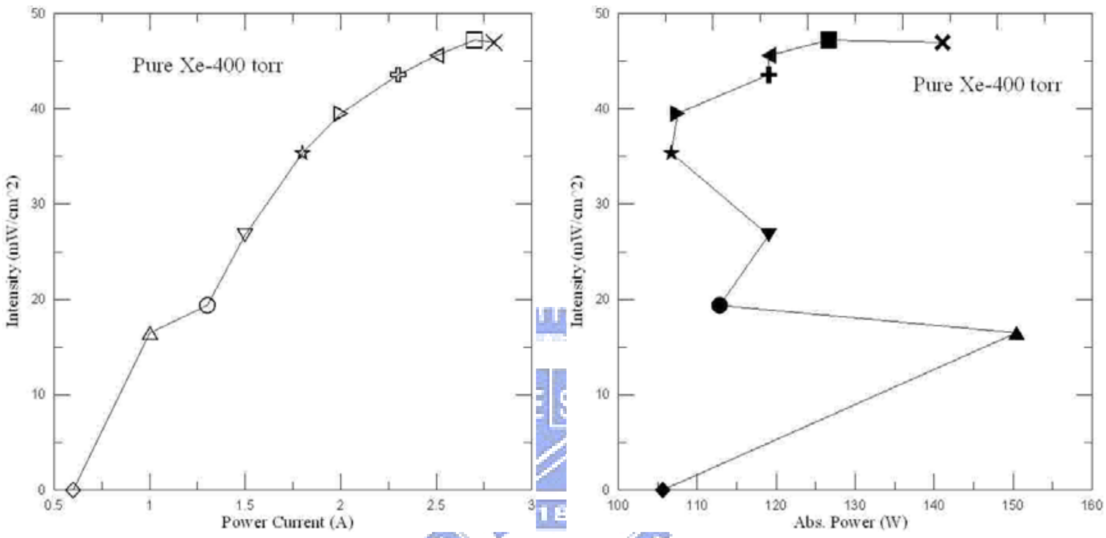
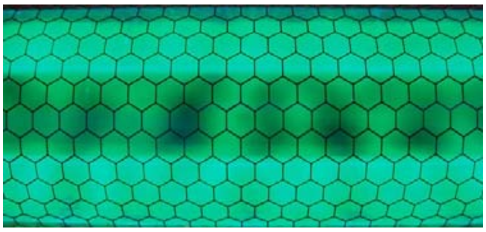
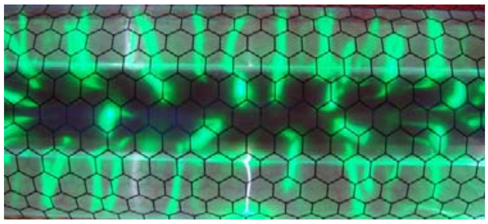
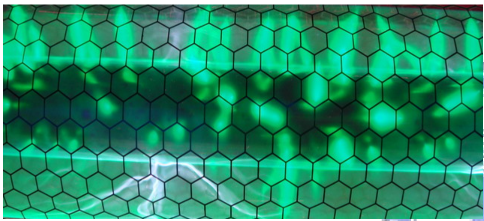


Figure 3.4 The light intensity change with different absorption power and power current in pure Xe and pressure is 400 torr.





(a) (b)



(c) (d)

Figure 3.5 The image of discharge situation with different pressure in mixture gas. The pressure is (a) 100, (b) 300, (c) 400 and (d) 500 torr.

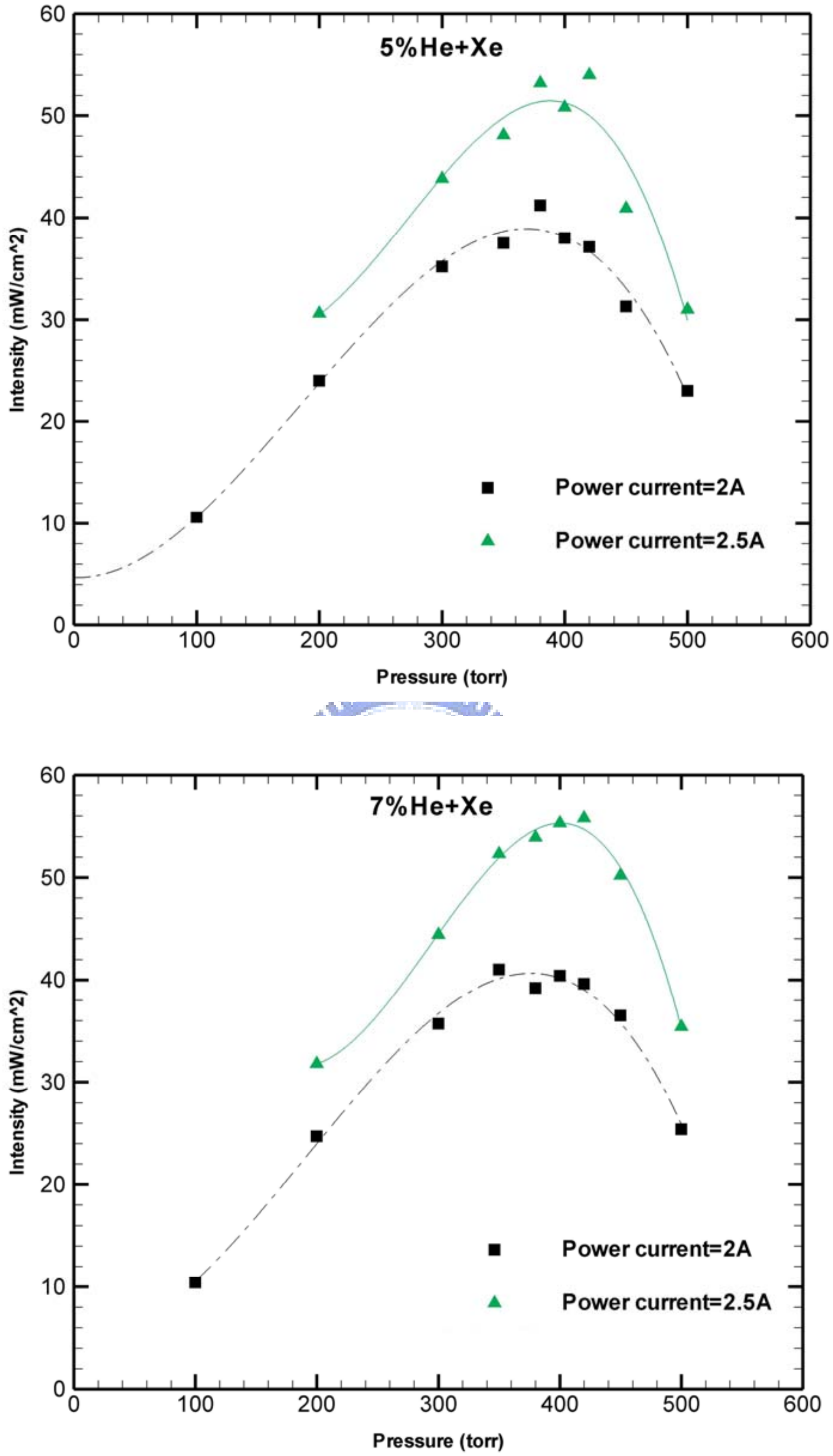


Figure 3.6 The light intensity change with different pressure in mixture gas.

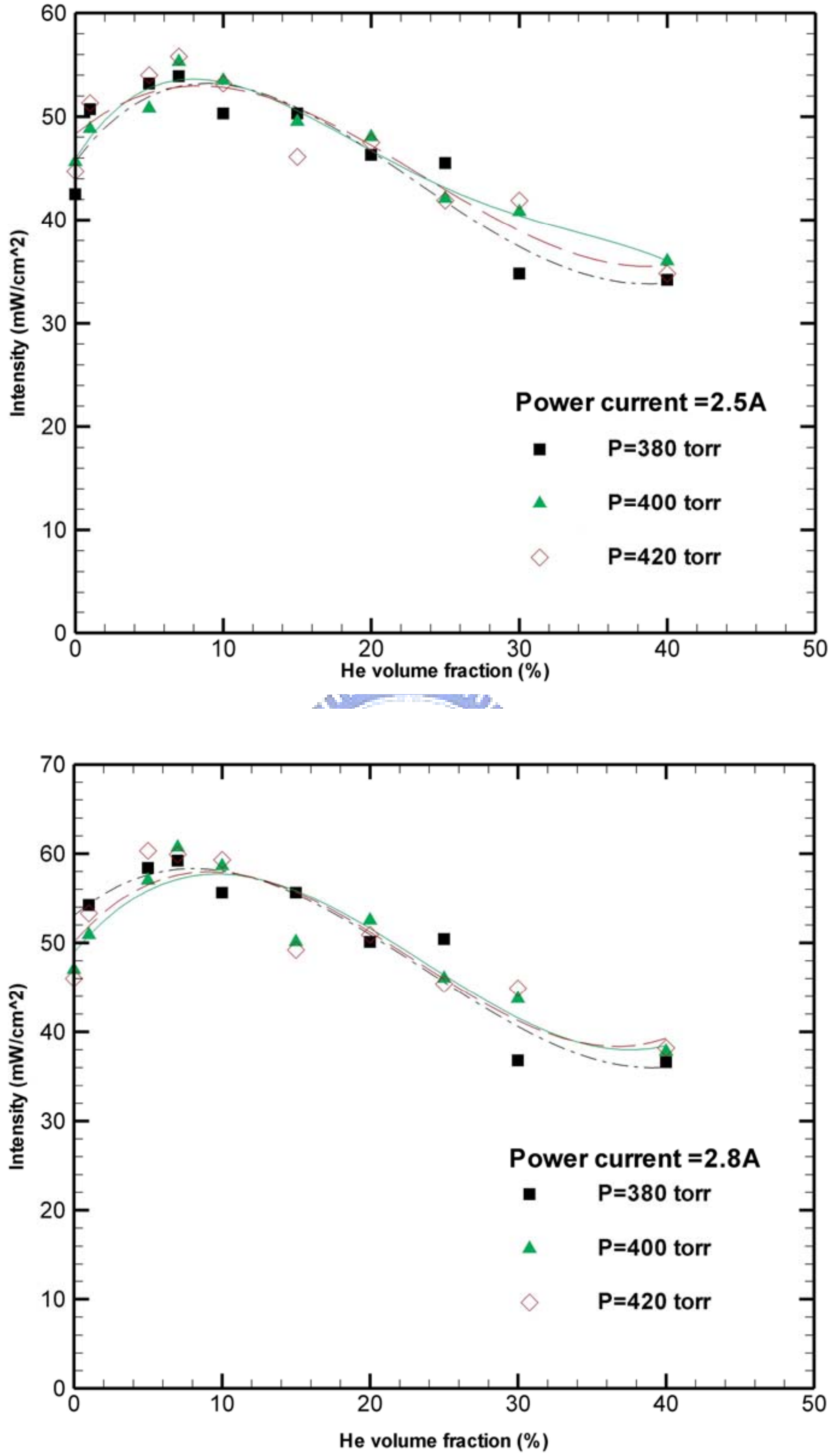


Figure 3.7 The light intensity change with different mixture fraction in mixture gas.

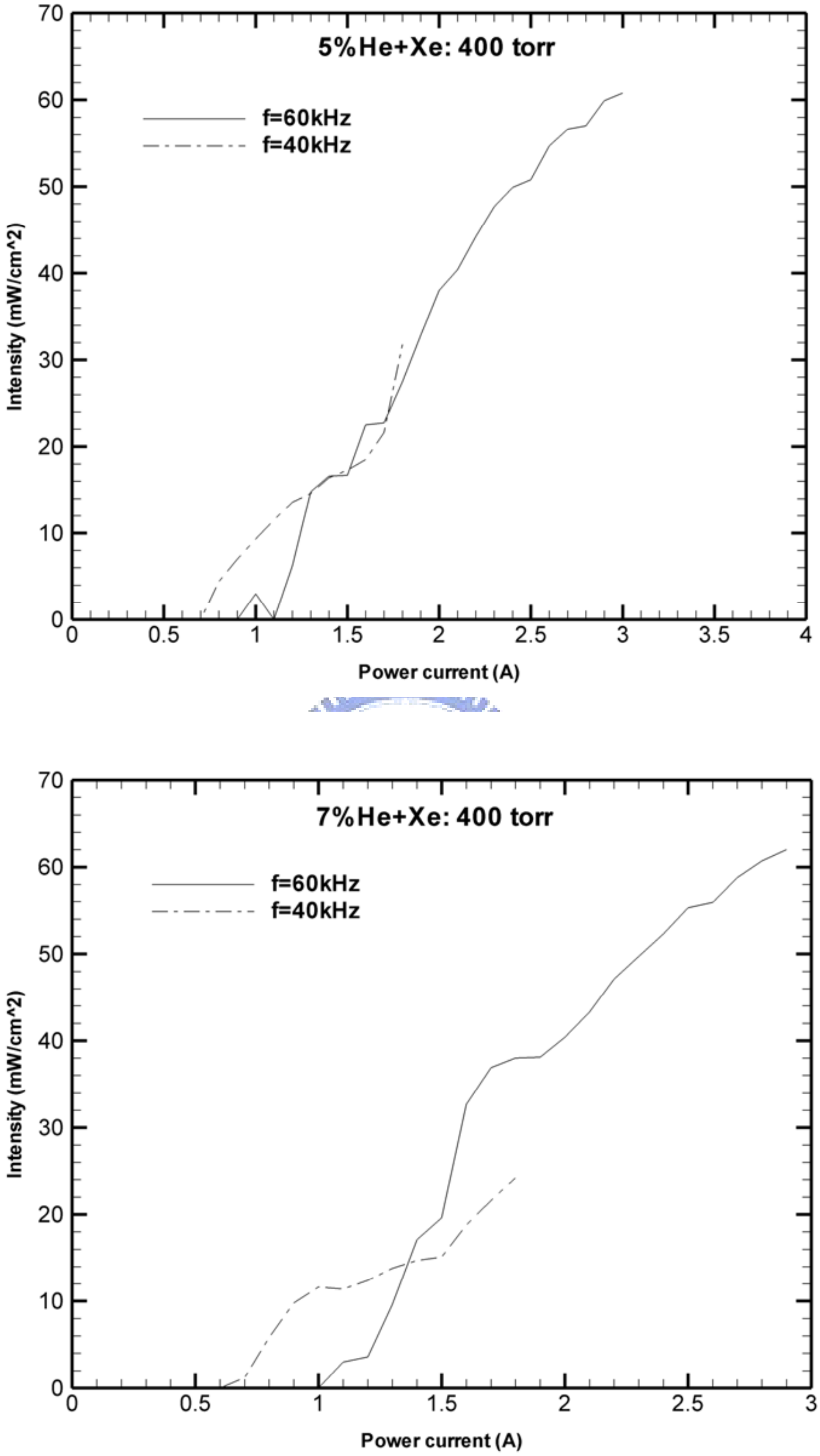


Figure 3.8 The light intensity change with different frequency in mixture gas and pressure is 400 torr.

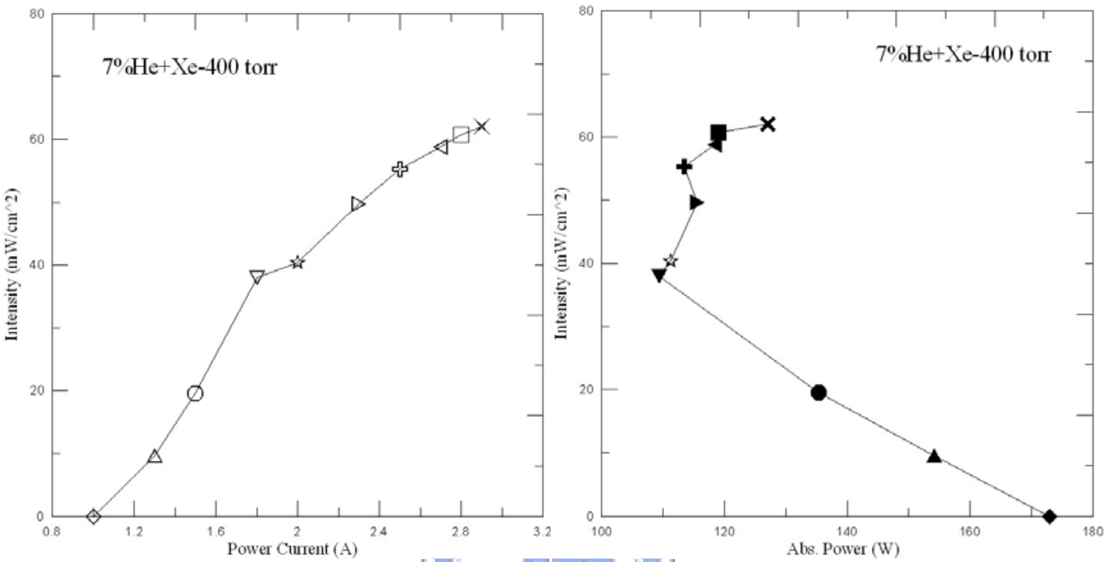


Figure 3.9 The light intensity change with different absorption power and power current in 7%He+Xe and pressure is 400 torr.