THE CORONA-DISCHARGE OF LIQUID AND GASEOUS HELIUM IN CRYO-PLASMA

SỰ PHÓNG ĐIỆN CỦA HELI LỎNG VÀ KHÍ TRONG PLASMA LẠNH

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Abstract - The use of a highly divergent field distribution (i.e. corona discharge or micro discharge) is essential in order to analyse different breakdown stages. In our work, we have measured the emission spectra and specially, the pressure broadening and shift of atomic lines and molecular bands. The experiments for point electrode (both negative and positive corona discharge) have been carried out at constant temperature, for gas at 300K; 150K; 10K; 6K and liquid at 4,2K; 5,1K in a wide range of pressure. The results obtained for pressure in the range 4,5-300 K will show the significant influence of pressure and perturber density on these phenomena.

Key words - Cryoplasma; corona discharge; helium; atomic spectrum; broadening; shift; interaction.

Tóm tắt - Việc sử dụng phân bố trường điện từ khác nhau ở mức cao (như phóng điện hay phóng điện nhỏ) thực sự cần thiết để phân tích những chu kỳ đánh thủng điện áp khác nhau. Bằng thí nghiệm thực hiện phóng điện trong Heli ở nhiệt độ thấp, phổ phát xạ được đo đạc và đặc biệt, độ rộng và độ dịch chuyển của phổ nguyên tử và phân tử được nghiên cứu. Với điện cực điểm – mặt phẳng (điện cực điểm có thể phóng điện dương hoặc âm), thí nghiệm được thực hiện ở các mức nhiệt độ xác định với trường áp suất thay đổi, thể khí ở 300K; 150K; 10K và 6K và thể lỏng ở 4,2K và 5,1K. Kết quả thu được trong khoảng từ 4,5 đến 300K sẽ cho chúng ta thấy ảnh hưởng quan trọng của áp suất và mật độ hạt nhiễu đối với những hiện tượng này.

Từ khóa - Plasma lạnh; phóng điện; heli; phổ nguyên tử; độ rộng; độ dịch chuyển phổ; thế tương tác.

1. Introduction

Liquid's electrical breakdown is a complex phenomenon that involves a succession of intercorrelated electronic, thermal, mechanical processes. These processes are called pre-breakdown phenomena. Investigations into such processes require applying very high electric fields. The use of a highly divergent field distribution (i.e. corona discharge or micro discharge) is essential in order to analyse the different breakdown stages. In order to obtain information about the important parameters that characterize non-equilibrium discharge plasma at both low and high pressures, we use a powerful tool named emission spectroscopy.

Spectroscopic observations of the light emitted by ionization gases can be used to determine conditions surrounding the emitted atoms or molecules. An ionization zone near a tip electrode is a source of a light emitted by the corona. Excited atoms interacting with environment and features of their spectra give information about density and temperature of a gas in the ionization zone.

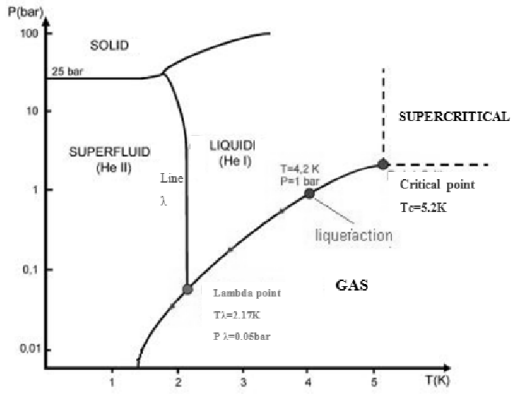


Figure 1. *Phase diagram of ^4He*

With helium, features of emission spectra of helium excited states and excimer molecules will be found by spectral analysis, which will be carried out on helium atomic lines and molecular bands from 300 down to 4,1 K. The reason for this choice is that according to the phase diagram of 4He (Figure 1), this substance can be found both in gas and liquid phases for different isotherms below the critical point of helium. We have chosen the isotherms at 4,2; 5,1; 6; 11; 150 and 300 K. The critical temperature of 4He is 5,2K at P=2,23bar, so upon variation of the pressure the data at 4,2 and 5,1 K will display the helium phase transition (Figure 1).

2. Experimental facilities

The experimental facilities used in our spectral investigations of corona discharges will be described in details below. The light emitted by the phenomena occurring in the high-field region near the electrode tip is analyzed by using a set comprising a plane spectrograph of Acton Research Corporation (ARC) and a multichannel optical detector. The 2D-CCDTKB-UV/AR detector is located directly in the exit plane of the spectrograph. One or two lenses are used to focus light onto the entrance slit of the spectrograph. The spectroscopic measuring device is shown schematically in Figure 2

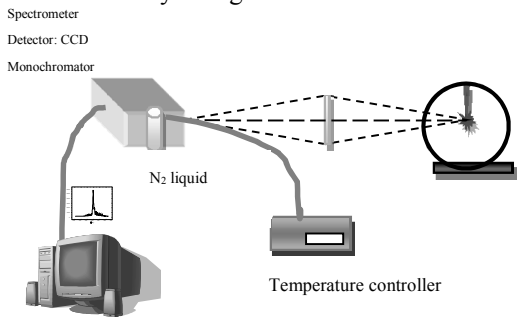


Figure 2. *Schematic diagram of the spectroscopy system*

Detecting the light exiting the spectrograph is powered

by a CCD-2D detection cooled with liquid nitrogen, called "LN/CCD" working at temperature of -120°C. The detector LN/CCD is controlled by a controller (ST-138, Princeton Instruments, Inc.), which allows controlling the acquisition and management of the detector. The 2D detector (model LN/CCD 512F&SB, Princeton Instruments, Inc.) includes a set of 512×512 pixels (size of each pixel 24×24μm) whose height and width are 12,3 mm in total. The background noise is low (≈1photoelectron/hour), which allows the acquisition of spectra without the need to correct the background noise even for long-term acquisitions. The spectra acquisition is controlled by a computer using the data acquisition software of "WinSpec".

The starting material was helium gas N60 (99,99990% pure, Air Liquide) which had an impurity concentration of about 0,1 ppm of oxygen. After the purification, the gas is liquefied in a cell housed in a cryostat. The temperature in the cell is measured by a germanium resistor and was fixed for each series of measurements. The measurements were carried out for different external pressures applied to the cell. The pressure increased until the spectral line was observed. Some measurements can be possible for the pressure 3,5MPa. The voltage was supplied by a stabilized DC power supply (Spellman model RHSR/20PN60) giving either positive or negative tip polarity. The stabilized dc voltage (up to 20 kV) was connected to the tungsten point electrode. The Tektronix TDS540 oscilloscope or the Keithley 610C current meter was connected to the plane electrode.

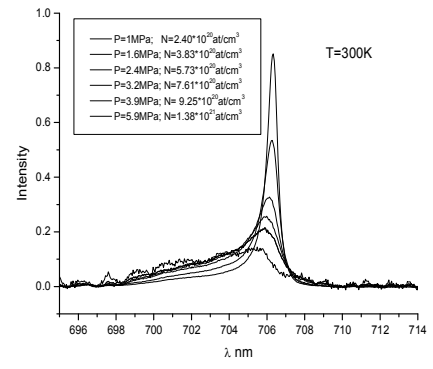
3. Results and discussion

All the lines identified at these temperatures are presented in Table 1. Emphasis will be placed on the 706nm, 728nm lines because they are the most intense lines and can be resolved on a very wide range of pressures.

Table 1. *Atomic lines of $^4He_2$ observed and identified at T=300K; 6K; 4,2K.*

λ(nm)	Upper state	Lower State	T=4,2-5,1K	T=6K-11K	T=150K-300K
388,86	3p ^{3}P	2s ^{3}S		389,15	388,7
396,47	4p ^{1}P	2s ^{1}S		396,73	
447,15	4d ^{3}D	2p ^{3}P		447,36	446,98
471,31	3s ^{3}S	2p ^{3}P	471	471,12	470,91
492,19	4d ^{1}D	2p ^{1}P	*490,6*	492,3	492,11
501,57	3p ^{1}P	2s ^{1}S	501,52	501,53	500,8
504,77	4s ^{1}S	2p ^{1}P		504,64	503,98
587,56	3d ^{3}D	2p ^{3}P	587,46	587,82	587,35
667,82	3d ^{1}D	2p ^{1}P		667,9	667,41
706,52	3s ^{3}S	2p ^{3}P	706,27	706,33	706,4
728,13	3s ^{1}S	2p ^{1}P	727,72	728	728,024
1083,02	2p ^{3}P	2s ^{3}S	1082,3	1082,76	1082,8

Within the framework of this paper, we'd like to introduce some of our results in HeI 706nm: In liquid helium 5,1K – 4,2K, regardless of the test pressure, the spectra are obtained for a current from 0,1 to 0,5μA, which corresponds to an average dissipated power about 0,5mW - 3mW. In gas phase (300-6K), the current is higher and the spectra are obtained for currents ranging from 20 to 50 μA and therefore a power of about 20 to 100mW according to applied pressure. However, the spectra for a given temperature and pressure data do not depend on the imposed power. The distribution of atomic lines and molecular bands is the same, although their amplitude increases with the current.



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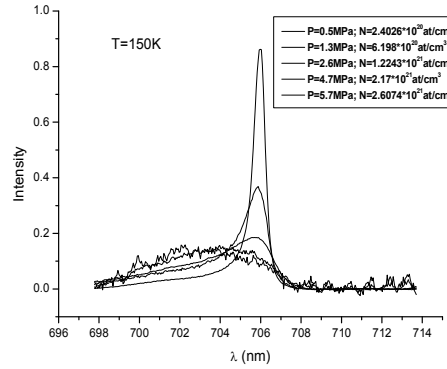

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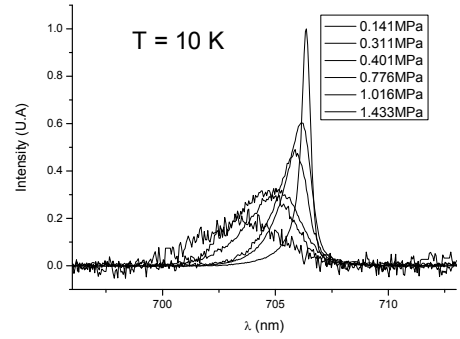

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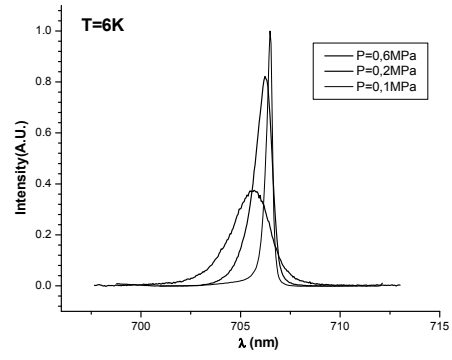


d)

Figure 3. *Variation of experimental profiles of 706,5nm ($3s^3S \rightarrow 2p^3P$) with pressure. Corona discharge in gas Helium at*

a) 300K b)150K c)10K and d)6K.

In gas phases, the atomic line at 706 nm ($3s^3S \rightarrow 2p^3P$ transition) being broadened and blue-shifted with increasing pressure. However, Figure 3 (a-b-c-d) shows the asymmetrical shape of the atomic line at 706m when pressure increases in gas from 6K to 300K. This asymmetry may be assigned to blue satellite bands at proximity of the atomic line. By increasing the pressure in the cell, the effect of the shoulder at 706,5nm is starting to be noticeable; this effect is dramatically illustrated by a sudden change in the slope of Figure 5. The line $3s^3S - 2p^3P$ at 706nm can be observed up to 5,9MPa.

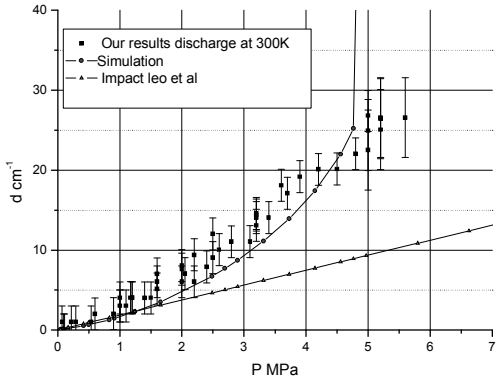


Figure 4. *Shift of the line λ=706,5nm as a function of pressure for T=300K*

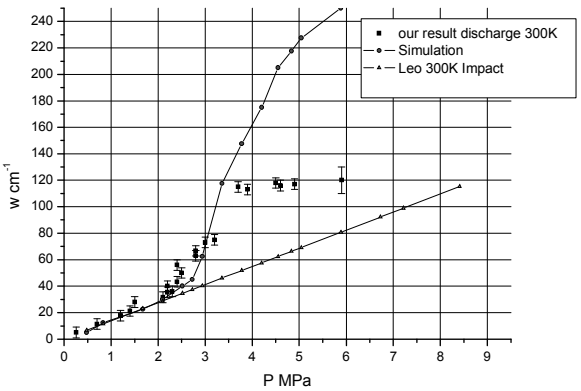
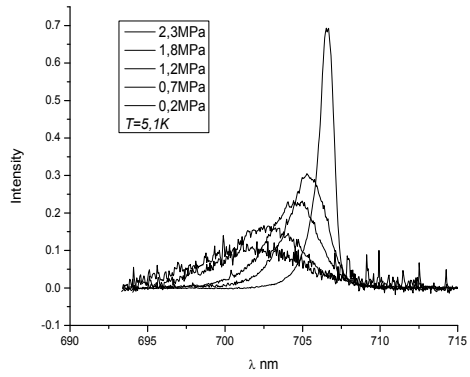


Figure 5. *Width of the line λ=706,5nm as a function of pressure for T=300K*

On the contrary, as can be seen in figures 6.a) and 6.b), the atomic lines of liquid He 706nm at 5,1K and 4,2K are broadened and blue-shifted with increases in pressure but with no significant changes in the symmetry of the line shape. On the other hand, the lines gradually disappear with increasing pressure. The $3s^3S$-$2p^3P$ line at 706nm can be observed up 3,5Mpa. The retained symmetric character of the line allowed us to quantify the width using the full width at half maximum (FWHM).

Therefore, the blue-shift in gas and liquid helium is pressure dependent. However, the fluorescent lines observed in liquid helium have a symmetric profile as opposed to high-density gas experiments.



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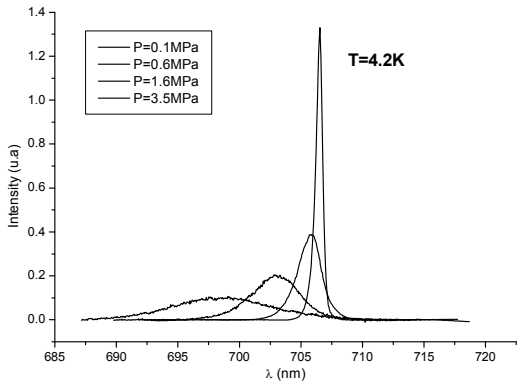

b)

Figure 6. *Variation of experimental profiles of 706,5nm ($3s^3S \rightarrow 2p^3P$) with pressure. Corona discharge in Helium liquid at a)5,1K and b)4,2K*

The temperatures T can then be calculated by knowing the densities N, assuming that the pressure in the ionization zone is equal to the applied pressure. The temperature rise in the ionized zone is 100 to 200K which is consistent with the temperature measured in a corona discharge in nitrogen by determining the rotational temperature of the second positive N_2 for a current of the order of 50μA.

With liquid He, the atomic lines remain strangely symmetrical. Since we observe no asymmetry blue wing that would let predict the use of the quasi-static approximation. So we can not apply the method used at 300K to deduce the density of the perturbers using the shift and the width of the 706nm line. Furthermore, the shift becomes linear as a function of the density whereas the width becomes proportional to $(N)^{1/2}$ [1].

The shift in Figure 4 we obtained is very close to that observed in superfluid helium excited by electron or proton bombardment. For example, Soley *et al.* [2] have made measurements on the line at 706,5nm excited by an electron beam at *T=1,75K*. They observed that this line was shifted to the blue with a shift as a function of pressure close to ours: 64,2cm^{-1}/MPa for Soley and 47,1cm^{-1}/MPa for our measurements. On the contrary, our results are consistent with those of Zimmermann [3] who recorded spectra produced by field emission in superfluid helium at 1,7K (Figure 7).

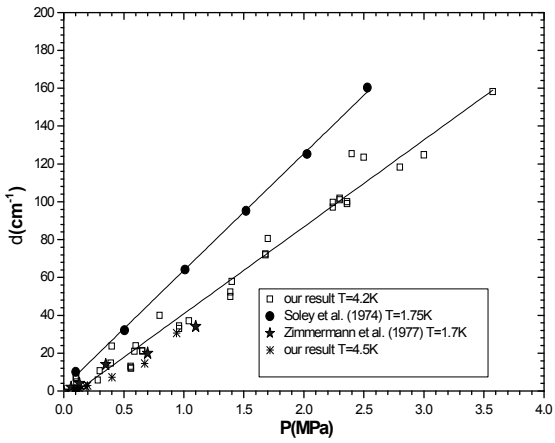

Figure 7. *Experimental shift of the line $3s^3S$- $2p^3P$ at 706,5 nm in liquid helium at 4,2 K; 4,5K and 1,7 K*

In all experimental work concerned with superfluid helium, the emitted light has never been interpreted as resulting from the formation of highly localized plasma (due to low bombardment current, it can be seen that the superfluid helium cannot be heated locally but from an excited atom or molecule surrounded by liquid.

In these experiments, they conclude that:

- The shift of the line (although important) is much lower than the expected one by considering liquid density affected by pressure.

- There is a difference in the shift between emission and absorption lines of the same line [4].

These conclusions have suggested the creation of a bubble around an atom or molecule, similar to the excited electronic bubble [5]. The bubble model has been developed for excimers in superfluid liquid He II at temperature 1,7K. The spectra recorded at T=4,2K shows that such phenomenon is possible in normal liquid helium also [6]. The formation of bubbles is explained as follows: after excitation, the outer electron interacts strongly with the surrounding atoms. This interaction causes a strong repulsion of the surrounding atoms and they are pushed away from the excited atom, or from excited molecular in a very short time [7] and creating a vacuum around He* and He^*_2. The excited atom is represented by a core He* and an outer electron.

The theoretical predictions on broadening and shift are shown in Figure 8 for the emission of 3^3S-2^3P in a pressure range from 0 to 2,5MPa using the bubble model of Steets et al. [8] and Hickman [9], [10]. The subsequent theoretical bubble model calculations appeared to provide a satisfactory agreement with the experiment [11].

By comparing the experimental values with the theoretical values derived from the bubble model, we observe that theoretical values are larger than our experimental displacement (Figure 8a), whereas the width values are the opposite (Figure 8b). It may be noted that our values of broadening are very close to those deduced from the bubble model at P<1,5MPa. If we plot the S/Δλ, the experimental ratio is far from the bubble model. (Figure 9).

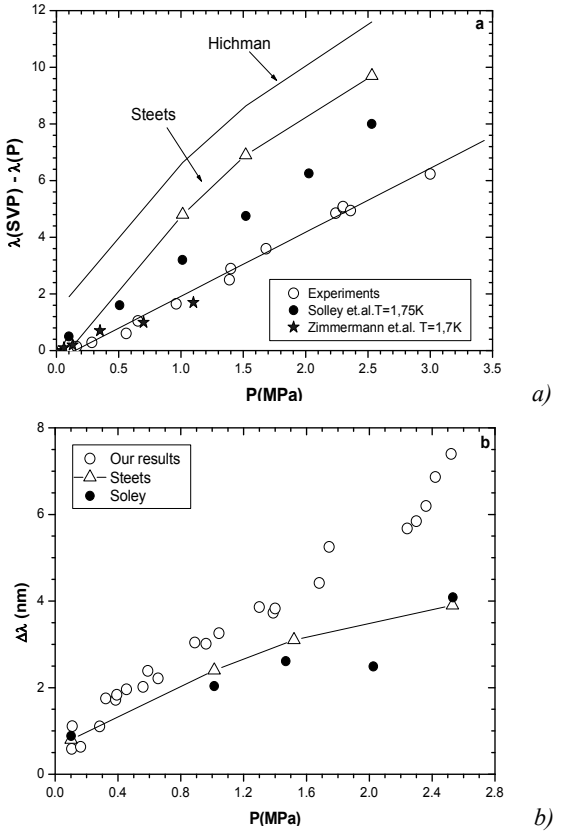

Figure 8. *Comparison between theoretical predictions of Steets et al. [12], of Hickman [13] and experimental results of Soley et al. [2] and our results for the shift (a) and width (b) of the line at 706nm*

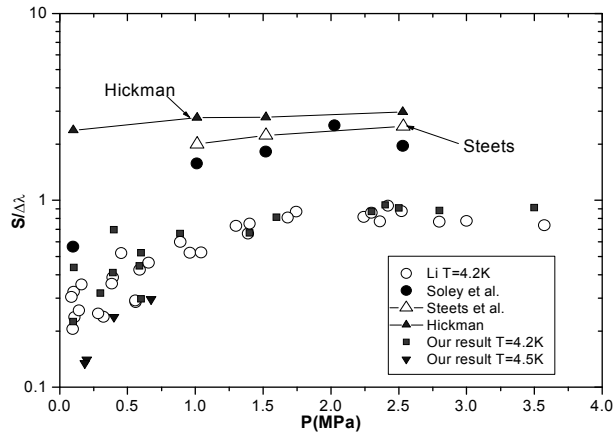

Figure 9. *Comparison of the ratio S/Δλ between the bubble theory and experimental results*

4. Conclusion

In gas phases, because of the low density, the shift of the line is very small but the spectral broadening is important due to frequent collisions with gas-phase He atoms. On the contrary, in the liquid phase, the blue-shift and line broadening are both large due to the higher helium density around He* [14]. The inter-atomic repulsion is the reason for the "blue" shift observed in spectroscopic experiments with gaseous and liquid He.

The asymmetric shape of spectral lines has been observed in He gas from 5K to 300K. The asymmetry of a

spectral line shape is described using "quasi-static" approach where perturbators are considered practically at rest. The quasi-static approximation can explain the intensity decrease of the spectral line upon the pressure increase.

Spectral analysis on liquid helium has raised a number of questions. In particular, the blue shift of the atomic lines at 4,2K, is very close to the shift in superfluid helium, and can not be explained by conventional potential interaction as Lennard-Jones potential. It is obvious that the *ab initio* potential becomes necessary as well as a proper quantum mechanical treatment of the collision for analysis of the line profile. The model of the pseudo bubble for the excited states can qualitatively explain the blue shift, but quantitatively, the difference remains significant. The reason for the particular behavior in liquid helium is certainly due to the formation of an ionization zone with a higher temperature T than 4,2K and a lower density than the liquid density but dense enough for the excited states to be stabilized in pseudo bubbles.

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