Luminescence from Liquid Helium Excited by Corona Discharges

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ABSTRACT

Liquid helium (LHe) at 4.2 K was electronically excited using a corona discharge for both negative and positive high voltages. The experiments were carried out for different pressures in the range from 0.1 to 10 MPa at constant temperature. The light emitted from the zone close to the tip was spectroscopically analyzed showing features from atoms and excimer helium. The shifts and widths of the observed lines and bands were found to depend on the applied hydrostatic pressure and on the tip polarity. Our analysis showed that classic pressure broadening theory cannot account for the observed widths and shifts rather than the presence of bubbles which surround single excited atoms and molecules. For positive tip polarities red shifted features distinct from pure He and He_2* were observed and tentatively assigned to "red satellites".

Index Terms —**Helium spectroscopy, corona discharge, excimer helium.**

1 INTRODUCTION

LIQUID helium is a fascinating substance with many peculiarities due to its highly quantum nature. A particular feature of liquid helium is its intense luminescence in the visible and near infrared spectral range. This luminescence has been observed from superfluid ^{4}He bombarded with energetic electrons [1-3], from liquid helium excited by a corona discharge [4-8] as well as from ^{4}He droplets excited by monochromatic synchrotron radiation [9-11]. After electronic excitation, Rydberg-type He atoms or excimer molecules are formed in liquid helium and a repulsive force between the Rydberg electron orbital and the surrounding ground state helium atoms is established. As a consequence the surrounding helium atoms are pushed away within a short time [12-13] creating a void around the excited atoms He^* and molecules He_2^*. This void is often referred to as a bubble and it has typical radii between 10-14 Å depending on the electron's orbital radius [14]. Bubbles of similar type are well known to enclose electrons in liquid and even dense gaseous helium [15-17]. Within the confinement of these bubbles the perturbation by surrounding ground state helium atoms is low and the electronic life time of the excited atoms or excimers is almost similarly long as for free species in the vacuum. The remaining perturbation by the ground state helium atoms surrounding the bubble is nevertheless strong enough to cause broadening and wavelength shifts of the atomic and molecular lines and the magnitude of the width and shift was found to depend on the applied pressure. The hydrostatic pressure was also found to affect the line intensity distribution of the rotational spectrum of the confined He_2^* as well as the total luminescence yield.

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Corona discharge represents a relatively simple and versatile way to produce and investigate electronic excitations and luminescence in liquid helium as it allows changes of pressure and density over a very wide range. However, the mechanisms involved in such a discharge in the liquid phase are complicated and involve electronic avalanches, energy transfer, formation of shock waves and bubbles, etc. Another complication is that the particle density in corona discharge is not uniform [18]: the region close to the tip has a high excitation density and thus resembles in many respects a plasma. It is a priori not clear whether under these conditions in the zone close to the tip the liquid state of helium is retained. To obtain a better understanding of these effects we have initiated a systematic spectroscopic investigation of liquid helium which is excited by corona discharges.

In this paper we report on the effect of the tip polarity on the spectra obtained in the liquid phase at 4.2 K as well as the effect of external hydrostatic pressure. We show that most spectral features are due to light emission from excited He atoms and excimers. The features are broadened and shifted depending on te hydrostatic pressure. Classical line broadening theory cannot explain the magnitudes of the line shifts and widths and we find that the most likely origin of the perturbation is the presence of bubbles around the emitting species similar to the observation by Dennis et al. [1] who used electron bombardment for the excitation of superfluid helium. We further show that additional red-shifted spectral features exist that cannot be explained by rotational line intensity distributions of thermalized excimer molecules. These features depend on the polarity of the corona discharge giving rise to a provisional assignment to 'red satellites' side-bands. The microscopic origin of these red satellites is presently unknown.

2 EXPERIMENTAL TECHNIQUES

Our experimental set up has been described elsewhere [4] and we only briefly mention its most important features. The liquid sample was produced from helium at the grade N 60 (Air Liquide) with an impurity concentration of about 0.1 ppm of oxygen. The gas was further purified by a series of traps that were filled with a mixture of molecular sieves (3-10 Å) and charcoal, activated under vacuum typically at 350°C for 3 days and finally immersed into liquid N_2. The liquid helium sample was obtained by condensing purified helium gas into a copper beryllium coaxial cell. The cell included a point electrode and had a characteristic impedance of 50 Ω when mounted into a cryostat and it also could withstand pressures up to 10 MPa. Before filling the cell was pumped to about 10^{-4} Pa using a turbo-molecular pump. Tungsten tips with a radius of 0.45µm and 2.5 µm were prepared by electrolytic etching. The electrode spacing was 8 mm. All metallic electrodes were supported by Macor insulators. In the cryostat the temperature of the liquid could be adjusted to 4.2 K at a constant pressure P. The high voltage from a stabilized dc power supply (Spellman RHSR/20PN60) was connected to the point electrode. Light emitted from the region close to the point electrode was analyzed by a spectrograph through a sapphire window. The spectrograph (Acton Research Corporation of 300 mm focal length) was equipped with 3 plane gratings: one with 150 gr./mm and two with 1200 gr./mm that were blazed at 750 nm and 300 nm, respectively. The 2D-CCDTKB-UV/AR detector was located directly in the exit plane of the spectrograph. In order to reduce the dark current, the detector was cooled to a temperature of 153 K (dark current < 1 electron/pixel/hour at 153 K. The instrumental broadening measured by recording the profiles of argon lines from a low pressure discharge lamp was $\Delta\lambda_{ins}$ = 0.12 nm for the 1200 gr./mm gratings.

3 EXPERIMENTAL RESULTS

3.1 GENERAL REMARKS

The light emitted from the corona region was collected and spectra in the range 500 - 1080 nm were recorded. Figure 1 shows a representative spectrum observed in LHe. Several atomic lines and molecular bands can be identified. These lines correspond to radiative transitions between excited states of He^* atoms and He_2^* excimer molecules.

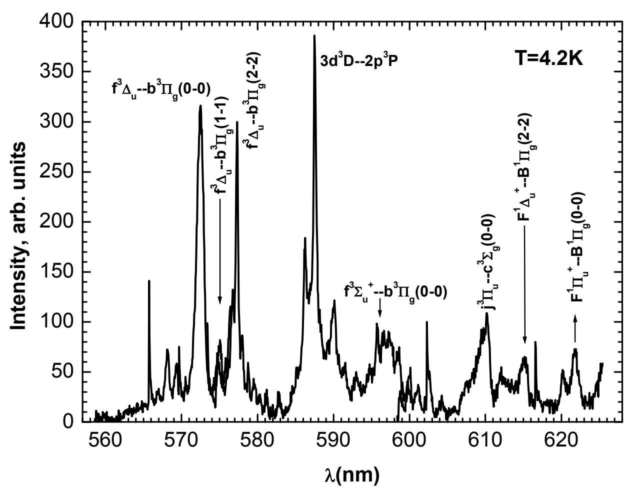

Figure 1. Overview spectrum recorded in the range 560-630 nm in LHe at 4.2 K, 0.1 MPa.

At low pressure the lines are sharp and their peak position match the atomic lines and molecular bands of helium from gas phase experiments. These lines are listed in Table 1.

A strong background continuum from 490 to 1100 nm appears in spectra at high pressures above P = 4.0 MPa. Moreover, the width of the lines increases with pressure and their relative intensity decreases. Figure 2 shows the atomic $3s^3S$ -> $2p^3P$ line at 706 nm being broadened and shifted with increasing pressure towards smaller wavelengths (blue shift). The relative intensity of the line decreases with pressure. No lines and bands can be observed in spectra if the pressure exceeds 5.0 MPa.

Owing to the relatively small mass of He atoms the excimer spectrum of He exhibits a spectrum with comparatively large rotational line separation which our spectrometer is able to resolve. Figure 3 shows that for low pressures rotational lines can indeed be observed allowing us to derive the population of upper rotational levels of the excited molecules. For the case that the intensities are governed by a Boltzmann distribution it is possible to assign a "rotational temperature".

Table 1. Transitions observed in liquid helium (T=4.2K, P=0.1MPa)

Atomic lines		Molecular bands	
λ(nm)	Upper-Lower	λ (nm)	Upper-Lower
492,19	4d ^1D-2p ^1P	462,24	$J^1\Delta_u$- $B^1\Pi_g$
587,56	3d ^3D-2p ^3P	464,95	$e^3\Pi_g$- $a^3\Sigma_u^+$
706,52	3s ^3S-2p ^3P	573,49	$f^3\Delta_u(v=0)$-$b^3\Pi_g(v=0)$
728,13	3s ^1S-2p ^1P	575	$f^3\Delta_u$ (v=1)- $b^3\Pi_g$ (v=1)
1083,02	2p ^3P-2s ^3S	577	$f^3\Delta_u$ (v=2)- $b^3\Pi_g$ (v=2)
		588,7	$f^3\Pi_u^-$- $b^3\Pi_g$
		639,6	$d^3\Sigma_u^+$- $b^3\Pi_g$
		659,55	$D^1\Sigma_u^+$- $B^1\Pi_g$
		913,61	$C^1\Sigma_g^+$- $A^1\Sigma_u^+$
		918,3	$c^3\Sigma_g^+$- $a^3\Sigma_u^+$

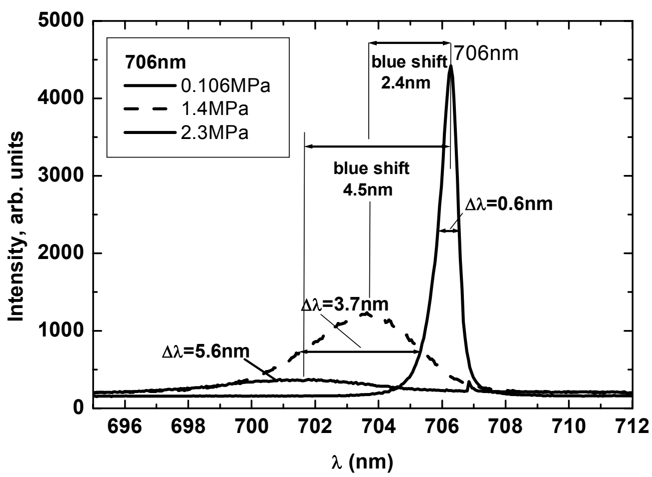

Figure 2. Intensity of the 706nm atomic line for different pressures. LHe at 4.2 K, I_{corona} = 0.3 μA, Power = 1 mW.

In gaseous discharges of molecular species the rotational temperature is often close to the thermodynamic temperature of the gas, however, exceptions especially for helium have been reported [19]. The different behavior of He is due to the excimer molecules being formed after the electronic excitation. Likewise, the rotational temperatures in the molecular bands observed in LHe or He droplets were by two orders of magnitude higher than the temperature of the liquid or the droplets [1].

In the following we will investigate the effect of hydrostatic pressure on the spectra and we will also investigate the effect of the polarity of the discharge. Increased hydrostatic pressure leads to broadening and shifts of the lines. The spectra recorded with positive tip polarity show distinct differences in the form of red-shifted features. These features were observed near atomic and molecular lines.

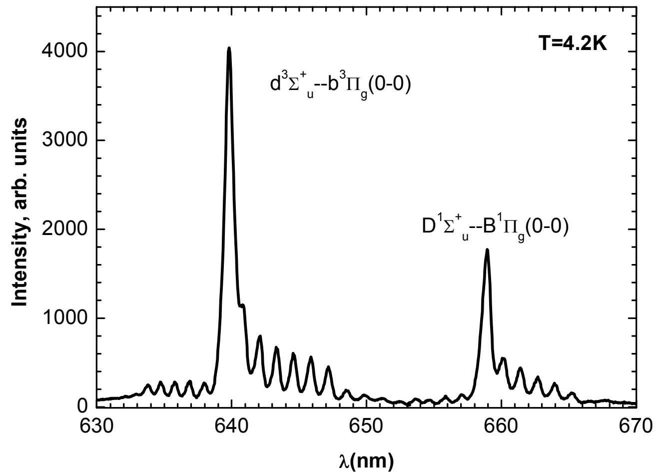

Figure 3. Molecular features in the spectrum of LHe at 4.2 K and 0.1 MPa showing rotational structure.

3.2 SPECTRA FROM NEGATIVE CORONA DISCHARGES

In this section, we show atomic and molecular spectra recorded with negative tip polarities. Figure 4 shows the same spectrum of the $3s^1S$ -> $2p^1P$ atomic transition at 706 nm as in Figure 2 but with normalized line intensity. In this way it can be seen that changes in pressure up to 1.4 MPa cause increased line widths, but with no significant changes in the symmetry of the line. Further increments in pressure up to 2.3 MPa produce slightly asymmetric line shapes. The retained symmetric character of the line allowed us to quantify the width using the magnitude of the full width at half maximum (fwhm). The magnitude of the shift was derived from the position of the maximum.

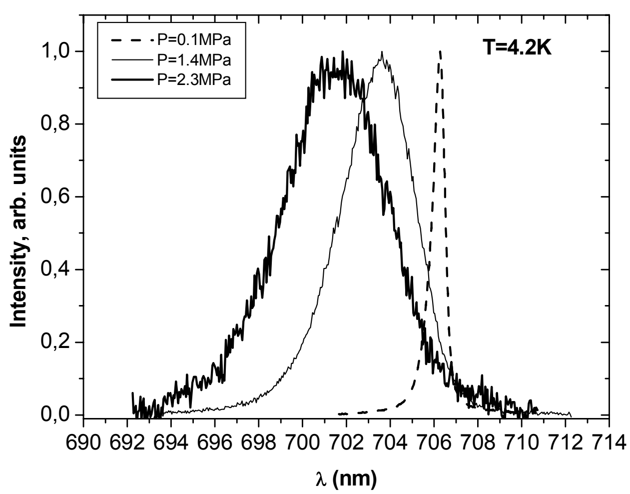

Figure 4. Normalized intensities of the 706nm atomic line for different pressures. LHe at 4.2 K.

In Figure 5 we show the dependence of the shift and width on the pressure. Both show a linear dependence on the pressure within the accuracy of our measurements. For high pressures the widths and the shifts have similar magnitudes which are remarkably distinct from the behavior of dense He gas at room temperature.

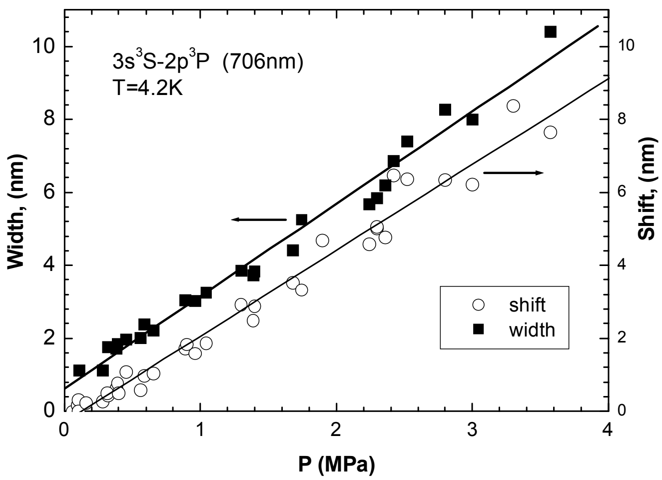

Figure 5. Shift and broadening of the 706nm atomic line vs. pressure. LHe at 4.2 K. Lines are linear fitting of the data curves comparing.

The molecular bands observed in our experiments with the negative corona show broadening and shifts with increasing pressure as well. The spectrum of the $d^3\Sigma^+_u$ - $b^3\Pi_g$ triplet transition at 640 nm is shown in Figure 6 for three different pressures.

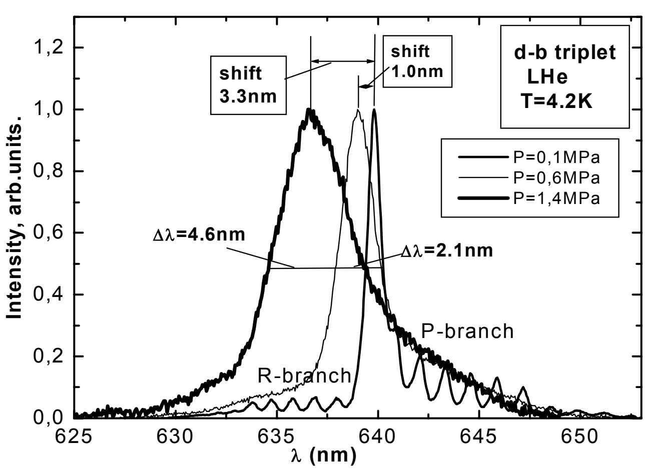

Figure 6. Transition $d^3\Sigma^+_u$ - $b^3\Pi_g$ in LHe at 4.2K for different pressures. The intensities are normalized.

As for the atomic lines the spectral features shift to shorter wavelengths with increasing pressure and become more broadened. The rotational structure of the band is resolved at 0.1 MPa and shows the R, Q, and P transitions due to changes of the rotational quantum number ΔN = 1, 0, -1, respectively. We note that the spacing of the Q-lines is too close to be resolved with our spectrometer. The widths of the R and P rotational lines are 0.7 nm at 0.1 MPa. With increased pressure the widths of the lines quickly becomes larger than the 1.3 nm wide spacing of the rotational P and R lines which makes the analysis very difficult. We note that the profile of the band recorded at 0.6 MPa is very similar to the spectrum reported by Dennis et al. who bombarded superfluid helium with high energetic electrons [1]. The shift of the Q-branch maximum with pressure is presented in Figure 7. Experimental data [2] obtained in superfluid He II at 1.7 K are also shown for comparison.

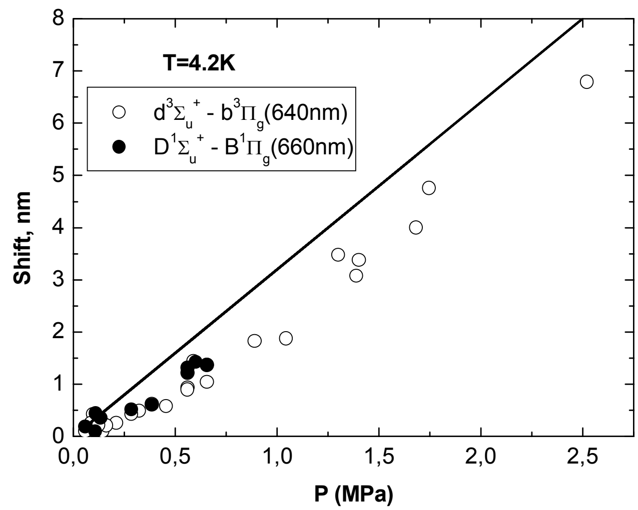

Figure 7. Shift of molecular bands for different pressures. The circles represent our experimental data whereas the line shows the shift reported by Soley and Fitzsimmons [2]. For pressures larger than 0.2 MPa the rotational lines overlap strongly and therefore the widths cannot be analysed.

3.3 SPECTRA FROM POSITIVE CORONA

Below, we show spectra recorded with positive tip polarity. These spectra show both atomic lines and molecular bands at 4.2 K over a wide range of pressures but very different line shapes compared to those obtained with negative tip polarities. Figures 8 and 9 show the atomic $1s3s^3S_1$ -> $1s2p^3P_{0,1,2}$ line at 706 nm and the $1s3s^1S_0$ -> $1s2p^1P_1$ transition at 728 nm at 4.2 K and 0.1 MPa. For negative tip polarity under same conditions these lines reveal no shift and no broadening (Figure 9), but in a positive corona discharge the lines clearly show a strongly asymmetric profile which can be fitted by a superposition of two Gaussian functions shifted from each other (Figure 10). The first, non-shifted feature has a width $\Delta\lambda_1$ = 0.706 nm. The second feature is shifted by 1.6 nm towards lower wavelengths and has a larger width $\Delta\lambda_2$ = 3.3 nm.

Figure 11 shows the molecular $d^3\Sigma^+_u$ - $b^3\Pi_g$ transition of He_2 using positive tip polarity. Similar to the atomic lines this band shows an asymmetric profile. In order to assess the degree of deviation we simulated the pure molecular $d^3\Sigma^+_u$ - $b^3\Pi_g$ transition (Figure 11) and we subtracted the simulated spectrum from the measured features. Figure 11 shows the resulting difference spectrum. This difference spectrum shows a clear asymmetry with regard to the pure molecular band and cannot be explained by broadening of by shifting of lines because P and R lines would be equally affected. The clear distinction from the well-known feature of pure He_2 molecules gives rise to assign the difference spectrum to a "red-satellite" of unknown origin.

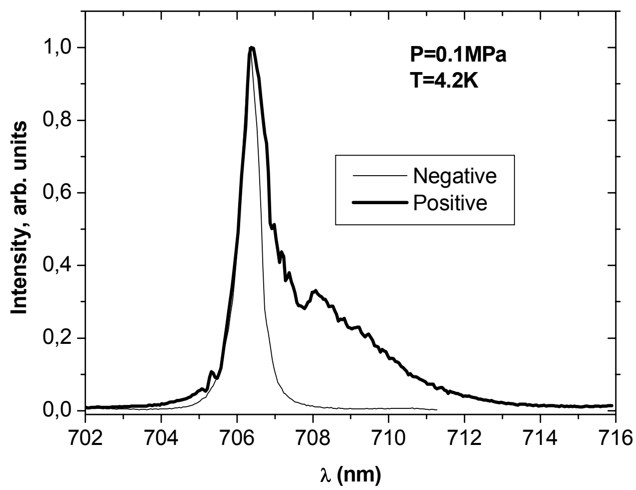

Figure 8. Atomic line of 706nm of negative and positive coronas in LHe, T = 4.2 K, P = 0.1 MPa.

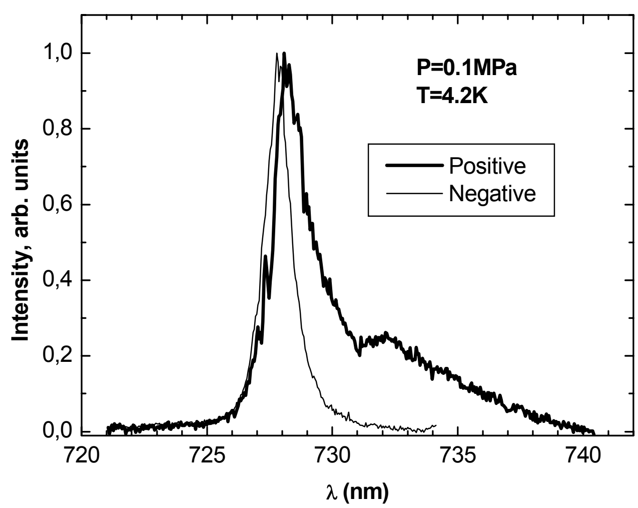

Figure 9. Atomic line at 728nm for negative and positive corona discharge excitation in LHe; T = 4.2 K, P = 0.1 MPa.

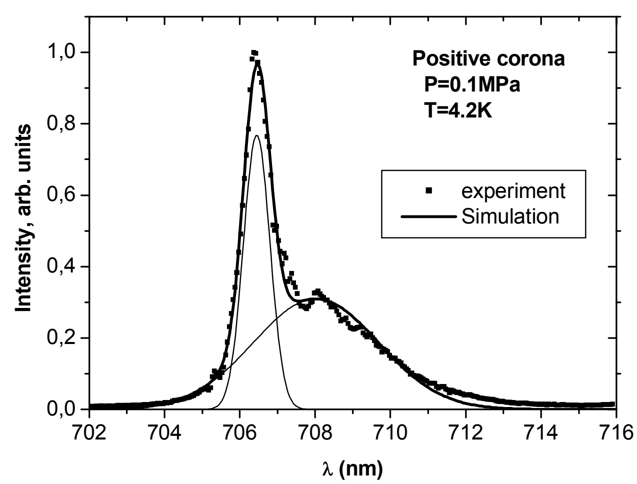

Figure 10. Atomic line of 706nm of positive corona in LHe simulated as superposition of two Gaussian profiles.

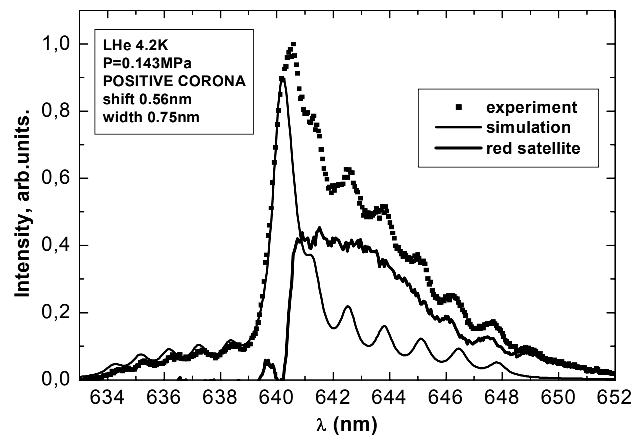

Figure 11. Rotational spectra of transition $d^3\Sigma^+_u$ - $b^3\Pi_g$ of He_2 in LHe, T= 4.2 K, P = 0.143 MPa.

4 DISCUSSION

The theory of the pressure broadening of spectral lines using so called "collision approach" is usually adopted to account for the line shapes observed in gaseous discharges [20]. Strictly speaking, the collision approach is only valid for low density gases but LHe may be an exception because it has the lowest density of all condensed matter and we therefore investigate its applicability.

The collision approach predicts symmetric Lorentzian line profiles with widths being proportional to the gas pressure. In helium gas the dependence of the line widths and shifts on the pressure are dominated by the repulsive interaction ($V(r) = C_{12}r^{-12}$) between an excited atom and surrounded atoms [21]. Within the so-called "limit of collision" for repulsive interaction, the expressions for the line broadening and the line shift are given by:

$$\Delta\lambda_{12} = 6.44\left(\frac{\lambda_{ul}^{2}}{2\pi c}\right)w^{9/11}\left|C_{12}\right|^{2/11}N$$

$$\Delta S_{12} = 0.922\left(\frac{\lambda_{ul}^{2}}{2\pi c}\right)w^{9/11}\left|C_{12}\right|^{2/11}N$$

Here λ_{ul} is the wavelength of the line in [m], $w=10^4*T^{0.5}$ is the relative velocity of the gas atoms with T being the gas temperature in [K], C_{12} is the repulsive Lennard-Jones parameter in [$m^{12}\ s^{-1}$] and N is the gas number density in [m^{-3}]. Regardless of the type of gas the collision approximation predicts a ratio between the shift and the width of 0.143 which is indeed close to what we find in our experiments with He gas at 300 K.

A consequence of the dominating repulsive interaction in He is that the lines are shifted towards shorter wavelengths. With increasing gas density the atomic He lines become more asymmetric in shape. This deviation from the initially symmetric blue-shifted lines can be accounted by the so-called "static approximation" [22]. We point out that when using positive tip polarity we did not observe any "red satellites" in the atomic line spectrum in gaseous Helium at 300 K.

In contrast to the gas phase we observe that in liquid helium the shift/width ratio is no longer constant and shows a distinct pressure dependence. For high pressures we find a value of about 0.8 for this ratio. For low pressures the shift/width ratio is lower but it never assumes a value of 0.143. The proximity of the shift/width ratio to the theoretical prediction may be taken as a measure for the applicability of the classical pressure broadening theory using the collision approach but we emphasise that deviations simply may indicate that other physical processes become relevant.

Previous theoretical and experimental studies on electron beam bombarded liquid helium have provided convincing evidence for the existence of microscopic cavities or "bubbles" having a diameter of about 1 nm which enclose excited atoms and excimer molecules within liquid helium [14, 23]. The origin of bubbles around an excited state of an atom or a molecule results from the balance between the repulsive interaction between the Rydberg electron and the ground state He atoms arising from the Pauli principle on the one hand and the pressure and the surface tension of liquid on the other hand. The shift of the spectral lines and their width depend primarily on the size of the bubble which in turn results from the balance between the repulsive force from the Rydberg electron and force on the bubble surface exerted by the hydrostatic pressure. Therefore the bubble size is pressure dependent.

It is a reasonable assumption that light emission from molecules enclosed in bubbles occurs in corona discharge-excited LHe as well and the similarity between our molecular spectra recorded at 0.6 MPa and 4.2 K with negative tip polarity and those reported by Dennis and coworkers [1] supports this interpretation. The pressure dependence seen in our spectra of LHe are therefore likely due to the perturbation exerted by the presence of bubbles. When the bubble size decreases with pressure the perturbation of the dominating upper level of the light emitting atom or molecule increases because the average distance to surrounding ground atoms becomes smaller. The line broadening theory which is valid for gaseous Helium cannot be applied anymore because it assumes the medium around the perturbed species to be homogeneous. This is no longer the case for a bubble. In Figure 12 we have also compared the measured widths of the 706 nm atomic line with the widths predicted by the "bubble" theory developed by Hickman et al. [14]. While there is agreement for small pressures deviations become significant at pressures larger than 1.0 MPa.

We note further that the bubble model of the spectral line shape predicts larger shifts than widths. The shift/width ratio calculated in [14] is close to 3 which is significantly larger than that predicted for "collision broadening" in gaseous discharges which can be taken as a major attribute of the spectrum of an excited atom inside a cavity in LHe.

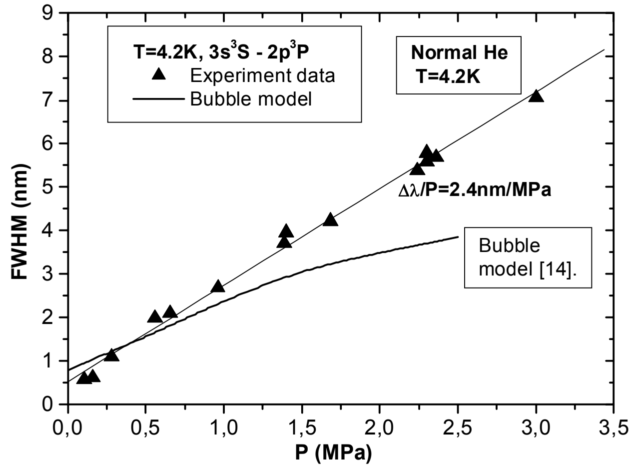

Figure 12. Full-Width-at-Half-Maximum (FWHM) of the 706 nm atomic line vs. pressure. LHe at 4.2 K.

4.1 RED SATELLITES IN NEGATIVE CORONA

A very small contribution of "red satellites" to molecular spectral features can be found in negative corona discharges as well. We have simulated the population distribution of the upper rotational levels for P = 0.1 MPa, where the rotational structure of the band is still clearly resolved (Figure 13). A Boltzmann distribution corresponds to a linear function in this semi-logarithmic plot. The plot shows that a Boltzmann distribution is only seen for large N. The "rotational temperature" derived from this distribution is 780 K which is significantly higher than the temperature of 4.2 K of the liquid. The populations derived from the P-branch intensities are larger than those from R-branch intensities. This fact can formally be interpreted by the existence of an additional source of radiation that contributes a side-band at larger wavelengths to the spectrum. A simulation of the d-b spectrum in Figure 14 shows the magnitude of the contribution of such a "red satellite" band as the difference between measured and calculated spectra.

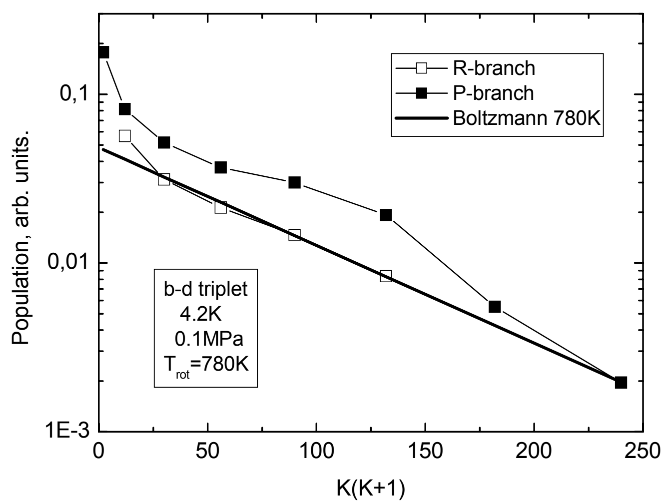

Figure 13. Relative population of the rotational levels of the upper term $d^3\Sigma^+_u$ - $b^3\Pi_g$ (idem to Figure 6) for negative corona.

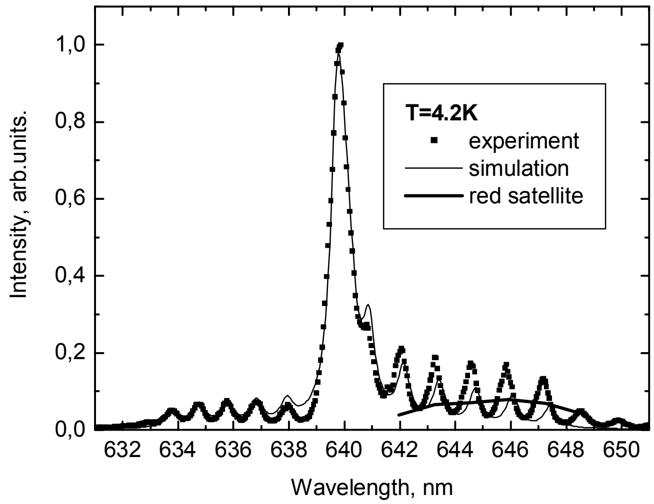

Figure 14. Rotational spectra of the $d^3\Sigma^+_u$ - $b^3\Pi_g$ transition: experiment, simulation and "red satellite" in LHe.

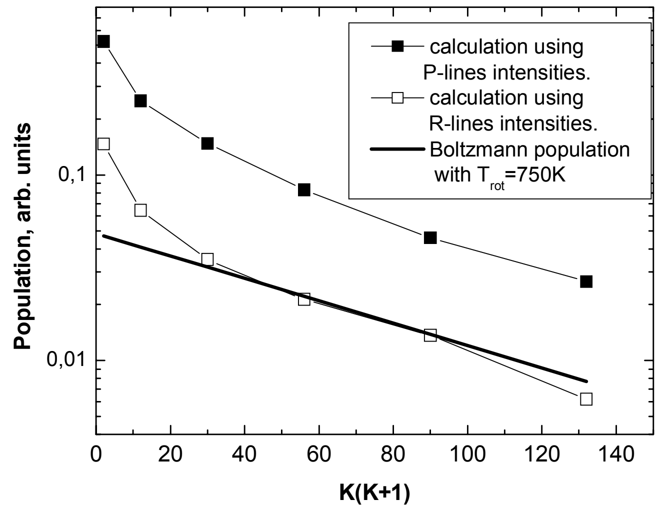

Figure 15. Relative population of the rotational levels of the upper term $d^3\Sigma^+_u$ - $b^3\Pi_g$ (idem to Figure 11) for positive corona.

4.2 RED SATELLITES IN POSITIVE CORONA

As stated before the intensities of the "red satellite" bands are much stronger in spectra recorded with positive corona discharges. A similar plot of the population distribution of the upper rotational levels to the one in Figure 14 is shown in Figure 15.

It is difficult to give a definite explanation for the nature of the red satellites. We have explicitly checked whether the "red satellite" stems from higher vibrational levels such as the v' = 1 -> v'' = 1 transition whose band head is located close to 642 nm and we found that this possibility has to be excluded. The strong tip polarity dependence suggests that charge carriers are involved. To make matters worse an explanation of the phenomenon of positive corona discharges has to the best of our knowledge not been reported yet, although its existence is well known. Clearly, a positive tip polarity raises the question as to how the electrons are released from the tip. It might be possible that free electrons are generated by entirely different processes, for instance, collisions with high energetic cosmic particles and that the role of the tip is only to generate high electric field strengths which eventually causes avalanches. The resulting excitation densities can therefore differ depending on the polarity. The asymmetric profile of the red satellite bands can be due to vibrational states of a van der Waals complex formed by the radiating atom or molecule and a helium gas atom in the ground state. We also recall the red-satellite features are only seen at 4.2 K, but not for gaseous He at 300 K.

Due to the many unknowns we cannot offer a conclusive explanation for the precise origin of the "red satellites".

5 CONCLUSION

We have shown that corona discharges are well-suited to excite visible luminescence of liquid helium at 4.2 K and to investigate its pressure dependence. The luminescence spectrum shows lines from transitions between electronically excited states of helium atoms and excimer molecules. For negative tip polarity and increasing hydrostatic pressure the lines become broader and shift towards shorter wavelengths.

For positive tip polarities a similar line spectrum can be observed, however, all lines are accompanied by red-shifted features. Also, the lines show much larger broadening than for negative tip polarities at similar pressure. Comparison between the P and R lines within molecular luminescence bands shows that the red-shifted features cannot originate from He excimer molecules and that they cannot be caused by a broadening process because P and R lines would then be equally affected. Therefore, the additional component in the spectrum is tentatively assigned to the contribution of an unknown "red-satellite". Since the underlying physical mechanisms of positive corona discharges are unknown it is not possible to further clarify the precise nature of the satellite spectrum on the basis of the available data.

Furthermore, we have investigated the dependence of the magnitudes of shift and broadening on the pressure for negative corona discharges. Classic line broadening theory predicts that these quantities are connected with each other such that the ratio between shift and width is a constant of 0.143. Our analysis of the spectral features of liquid He at 4.2 K reveals a higher ratio indicating that classic pressure broadening theory cannot be applied. We presume that the emitting atoms and molecules are localized in bubbles similar to the observations by Dennis et al., Soley and Fitzsimmons [1, 2]. The similarity between our spectral features and their reported spectral data supports our interpretation.

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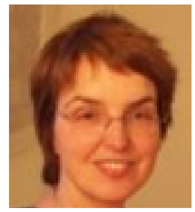

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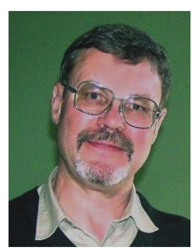

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