The Influence of Molecular Transitions on the Spectrum of the Helium Line at $\lambda = 447.15$ nm in a Low-Pressure Discharge

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Z. Naturforsch. 34a, 529-532 (1979); received March 10, 1979

In a linear pulsed discharge of helium the spectrum of the atomic transition 4^3D , $F \rightarrow 2^3P$ at $\lambda = 447.15$ nm is measured time resolved by a multichannel analyzer. Pronounced structures of the line profile, reported by other authors could not be verified and helium molecular lines did not occur in the parameter range of this discharge below 100 Pa. The measured influence of spurious hydrogen might suggest the interpretation of the recently observed structures as being due to hydrogen molecular transitions.

Introduction

The investigation of fluctuating electric fields in plasmas by means of spectroscopic methods has been extensively studied during the last ten years. Because of their special suitability the helium line 4^3D , $F \rightarrow 2^3P$ at $\lambda = 447.15$ nm has been used in many experiments. The intensity of the plasma satellites, appearing as a response to fluctuating electric fields is very small, however, and the probability of misinterpreting structures in the line profile, actually stemming from impurities, is rather high.

Recently Drawin and Ramette [1] have published measurements of the 447.15 nm line in helium, that show a large number of peaks in the line profile, which have been interpreted as being due to optical transitions of the helium molecule. Since earlier measurements of the same authors [2] under nearly the same discharge conditions did not show these irregularities in the line profile we repeated the experiment in order to clarify these discrepancies.

Experiment

The experimental set-up is shown in Figure 1. We used discharge tubes of different lengths (600 mm and 400 mm) with an inner diameter of 25 mm and different forms of Al-electrodes. The use of various capacitors allowed to change the total discharge time. The light from the plasma was observed as well end-on as side-on via lenses and apertures and was given on the entrance slit

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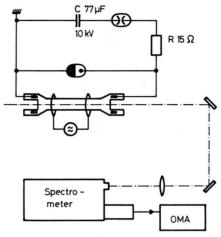


Fig. 1. Experimental set-up.

of a f=1 m spectrometer (1200 gr/mm). The exit slit of the spectrometer was replaced by the target of an optical multichannel analyzer. With its 500 channels we obtained an effective apparatus broadening of 0.025 nm (FWHM).

The capacitor bank is discharged through a current limiting resistor into the plasma by a spark gap.

The spectrum of the plasma can be monitored at an adjusted time during a time interval of 1 μ s or 3 μ s, respectively chosen for this experiment.

In addition to this pulsed plasma we also produced a high frequency discharge by capacitive coupling rf-power into the same gas tube ($f \approx 30 \text{ MHz}$, $p \approx 50 \text{ W}$).

Measurements

Figure 2 shows the profile of the helium atomic line at 447.15 nm at a gas pressure of 133 Pa.



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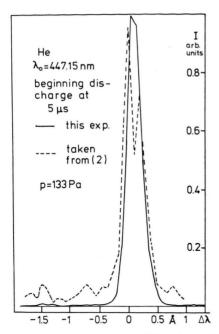


Fig. 2. Measured profile of HeI at $\lambda=447.15$ nm representing 80 individual shots.

This profile represents the average of 80 individual shots. The time of exposure was 3 μ s for every shot at the very beginning of the discharge.

As molecular lines of He₂ are expected to originate predominantly at the wall region of the plasma [3], the measurements have been done side-on. The error bar is — because of the large number of measurements — very small and lies within drawing accuracy.

In the same figure a profile of the 447.15 nm line reported by Drawin and Ramette [1] is shown. It represents the measurement at 10 μs at the beginning discharge at 133 Pa, too. Contrary to Drawin's measurement we do not see any significant structure though the reproducibility of the plasma parameters is very good.

The electron density of $N_{\rm e}=2\times 10^{19}~{\rm m}^{-3}$ taken from the weak forbidden component at $\varDelta\lambda=-1.5~{\rm \AA}$ is in accordance with the data from double probe density measurements [4] at the same time of the discharge.

Since Drawin has interpreted his strongly structured profiles as being due to optical transitions of the He₂ molecule, we looked for the intense $e^3\Pi_{\rm g} \to a^3\Sigma_{\rm u}^+$ band of He₂ at 465.0 nm.

In the pulsed discharge we could hardly detect this molecular band even when we opened the entrance slit of the spectrometer to 1 mm and integrated the light during the whole time of discharge (up to 300 µs).

Using the stationary highfrequency discharge we were able to monitor He₂-transitions as shown in Figure 3.

The variation of the intensity of the relatively strong band at 465.0 nm and the intensity of the atomic transition at 447.15 nm is demonstrated in Figure 4.

At pressures below about 100 Pa the band intensity is very small; it exceeds the intensity of the atomic line at pressures above 400 Pa. The maximum of the 447.15 nm intensity is indicating the formation of molecules at the expense of excited atomic states.

In this same highfrequency discharge the occurence of molecular helium transitions in the very neighbourhood of the 447.15 nm line (it is the $4^3\Sigma_{\rm u}{}^+ \to 2^3\Pi_{\rm g}$ band) is barely indicated at best as can be seen in Figure 5. The atomic line had to be excluded from recording (by placing the light

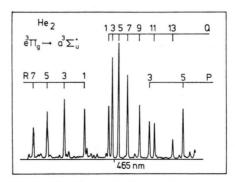


Fig. 3. The $\mathrm{He_2}$ molecular band around 465 nm emitted from a rf-discharge.

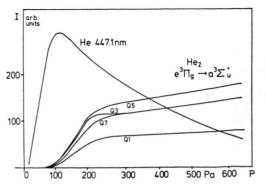


Fig. 4. Intensity of atomic and molecular transitions of helium as a function of gas pressure.

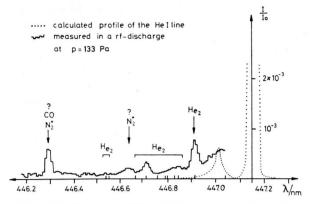


Fig. 5. The blue wing of the 447.15 nm helium line emitted from a rf-discharge.

outside the analyzer target) because of their high intensity. The rising background intensity towards the atomic line stems from the extreme over-exposure of the last channels of the analyzer target. The position and intensity of the atomic line and the forbidden component are indicated by the dotted line. Eventually appearing He₂ transitions reach an intensity of 5×10^{-4} of the allowed atomic line at best. These transitions are marked by short lines at the bottom of the figure, the wavelengths are taken from [5].

Discussion

Repeating the measurements of (1) at nearly the same discharge conditions we found, that He2 transitions do not affect the 43D, $F \rightarrow 23P$ line of atomic helium in this linear pulsed discharge at pressures below about 100 Pa. We conclude this from the fact, that reproducible structures near the 447.15 nm line can not be found and that strong molecular bands of the He2 molecule at $\lambda = 465 \text{ nm}$ can only be detected under extreme exposure conditions (1 mm slit width, very long integration time). In the discharge in which molecular transitions in the $e^3\Pi_g \rightarrow a^3\Sigma_u^+$ band occur, He₂ lines near the 447.15 nm atomic transition are of negligible importance below 100 Pa. Therefore the attempt of explaining the measured structures as being due to molecular helium lines seems to be doubtful.

There is a different explanation, however, that might be more adequate under certain assumptions.

When we used a discharge tube that had been operated in hydrogen before, the molecular spectrum of helium at 465 nm was completely suppressed and a strong hydrogen band spectrum appeared.

The wall contamination and the adsorbed hydrogen in the electrodes could only be removed by several hundred shots at the maximum current available.

Figure 6 shows the measured helium profile of [1] in the beginning discharge at $10~\mu s$ in a logarithmic plot. In addition a calculated hydrogen molecular spectrum is shown. Position and intensities of the molecular lines are taken from Dieke [5] and apparatus broadening has been assumed. The absolute intensity has an arbitrary value.

A comparison between measured and calculated spectrum indicates, that nearly all structures in the experimental profile can be explained by hydrogen molecular lines, provided the discharge tube has been in contact with hydrogen before.

In summary it may be expected that in this linear pulsed discharge at pressures below 100 Pa helium molecular lines are of vanishing importance as far as their influence in plasma satellite studies of the 447.15 nm atomic line of helium is concerned.

The influence of spurious hydrogen in the helium discharge however, should carefully be taken into account.

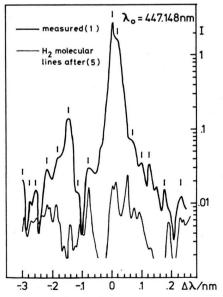


Fig. 6. Measured line profile of (1) in comparison to a calculated hydrogen molecular spectrum.

Acknowledgements

This investigation is part of the joint efforts within the Sonderforschungsbereich 162 Plasmaphysik Bochum-Jülich financed by the Deutsche Forschungsgemeinschaft. We are grateful to Prof. Dr. H. Schlüter for valuable discussion. The help of P. Andrzejewski and J. vom Scheidt is gratefully acknowledged.

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