VACUUM ULTRAVIOLET SPECTROSCOPY OF MICROHOLLOW CATHODE DISCHARGE PLASMAS

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1. Abstract

Discharge plasmas generated in hollow cathode configurations with hole sizes of 0.1 – 0.25 mm in pure high-pressure Ne and He (up to atmospheric pressure) were found to be efficient sources of Ne_2^* and He_2^* excimer radiation. Emission spectroscopy revealed strong emissions which were identified as the second and first continua of respectively the Ne_2^* excimer (75 – 88 nm) and the He_2^* excimer (60 – 90 nm). Admixtures of small amounts of H_2 (up to at most 0.1%) to Ne resulted in suppression of the excimer radiation and the emission of intense, monochromatic H Lyman-α and Lyman-β radiation at 121.6 nm and 102.5 nm, respectively. High-pressure He discharge plasmas with trace amounts of H_2, N_2, and O_2 also results in the emission of monochromatic atomic H, N, and O lines in the range from 95 – 125 nm in addition to the He_2^* excimer radiation. The sources of these intense monochromatic atomic line emissions are believed to be near-resonant energy transfer processes between the molecular gases and the rare gas excimers and/or metastables generated in the discharge plasma. Time-resolved fluorescence spectroscopy has been used to elucidate the microscopic details of these energy transfer processes.

2. Introduction

Hollow cathode (HC) discharges consist of a metallic cathode with hole, an arbitrarily shaped metallic anode, and an insulating ceramic in between. HC discharges show several modes of operation as a function of gas pressure p, hole diameter D, cathode-anode separation d, and discharge current I. At values of the product $p \cdot D$ below 10 $Torr \cdot cm$ and low currents (below 1 mA), a normal glow discharge develops along the path of the vacuum electric field. As the current increases, a transition to the hollow cathode mode occurs, in which the ionization is concentrated along the axis of the discharge and the discharge is sustained by energetic "pendulum electrons" [1,2]. If the current is increased further, an abnormal glow discharge develops. The hole diameter D is inversely proportional to the pressure up to about 10 Torr/D (in cm) for noble gases and N_2 [1-3], so that atmospheric-pressure operation requires a hole diameter of the order of 100 µm (microhollow cathode or MHC discharge).

HC and MHC discharges have been used extensively for the generation of non-coherent UV and VUV excimer radiation using either pure rare gas or rare gas - halide mixtures [2,3]. Rare gas atoms have a ^1S_o electronic ground state. The lowest excited-states result from the promotion of a (np) valance electron to the (n+1)s-level (n=2,3,4,5 for Ne, Ar, Kr, Xe) leading to four "P-states", two of which are metastable, while the other two states decay to the ground state via dipole-allowed transitions. Excimer molecules are formed via three-body collisions involving a metastable rare gas atom and two ground state atoms (and via other collisional interactions between electrons and atomic and molecular ions), if (i) there is a sufficiently large number of electrons with energies above the threshold for the formation of metastable rare gas atoms, and if (ii) the pressure is high enough in order to have a sufficiently high rate of three-body collisions. Rare gas excimer emission spectra are dominated by transitions from the lowest lying bound $^1\Sigma_u$ and $^3\Sigma_u$ excimer states to the repulsive ground state (second continuum) [3,4] with peak emissions at 170 nm (Xe), 145 nm (Kr), 130 nm (Ar), 84 nm (Ne), and 75 nm (He). Spectroscopy of the Ne_2^* and He_2^* excimers requires a specially designed "open" MHC discharge source [4] connected directly to a VUV monochromator, since no material is transparent below 105 nm.

3. Experimental Details

The electrodes of our MHC discharge (fig. 1) are made of 0.1 mm thick molybdenum foils separated by a 0.25 mm spacer of mica with a hole of typically 0.05 - 0.2 mm diameter in the cathode, the dielectric, and in the anode. Supply voltages V_o are typically 400 to 700 V and sustaining (discharge) voltages $V = R_{CVR} \cdot I_{DIS}$ are in the range of 200 - 300 V depending on the gas, the pressure, and the actual geometry of the MHC discharge. Discharge currents I_{DIS} vary between 1 - 10 mA. The circuit includes a resistor R_{CVR} which allows us to monitor the discharge current directly on an oscilloscope along with the discharge sustaining voltage V_{DIS}. We can also operate the MHC discharge in a pulsed dc mode with frequencies up to tens of kHz, pulse lengths from 100 ns to 1 ms and variable pulse separation and duty cycle using a versatile pulse generator.

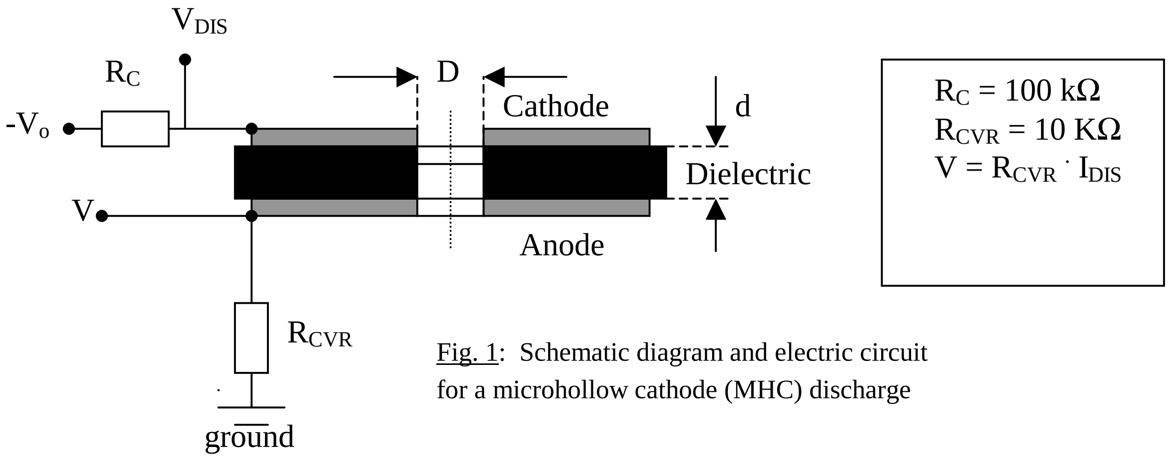

Fig. 1: Schematic diagram and electric circuit for a microhollow cathode (MHC) discharge

Measurements were carried out in high-pressure Ne or He (several hundred Torr) with small admixtures of H_2 (up to 2 Torr) using the MHC discharge source in an "open" mode, i.e. mounted directly to the entrance slit of a Minuteman 302-V 0.2 m VUV monochromator (wavelength range 50 - 250 nm, reciprocal linear dispersion of 4 nm/mm) or in a “closed” mode where the MHC discharge is sealed by a LiF window and only radiation of wavelengths longer than 105 nm (i.e. only the atomic resonance emissions) can be detected. In the “open” mode, the Ne and He radiation from the MHC discharge (60 -90 nm) enters the VUV monochromator through a 200 μm pinhole between the discharge region and the monochromator, which reduces the gas load from the high-pressure discharge. into the separately pumped monochromator and detector regions. The VUV photon were detected by a channel electron multiplier or a VUV photomultiplier tube connected to a standard pulse counting system.

4. Results and Discussion

4.1 Neon MHC Discharge Plasmas

The first experiments were carried out with a MHC discharge in high-pressure Ne (up to 760 Torr) either pure or with small admixtures of H_2 and N_2 (up to about 2 Torr). Fig. 2 shows the emission spectrum from a 740 Torr pure Ne MHC discharge in the spectral range from 70 - 95 nm. The emission feature between 73 and 78 nm corresponds to the Ne resonance lines at 73.5 and 74.3 nm and the Ne_2^* excimer first continuum. The broad emission feature in the region 76-88 nm is attributed to the second continuum of the Ne_2^* excimer emission. Fig 3 shows an emission spectrum between 70 and 125 nm from the same MHC discharge operated in a mixture of Ne (740 Torr) and H_2 (1 Torr). There are essentially no emissions in the region of the Ne resonance lines and the Ne_2^* excimer. The spectrum is totally dominated by two intense atomic lines at 121.6 nm and 102.5 nm which coincide with the Lyman-α (121.6.nm) and Lyman-β (102.5 nm) lines of atomic hydrogen. The intensity of these two atomic line emission is orders of magnitude higher than what one expects from a conventional discharge containing 1 Torr H_2 [4,5].

Wieser et al. [5] recently reported a similar emission of intense H Lyman-α light from a mixture of high-pressure Ne (>200 Torr) with a trace of H_2 in a gas cell excited by high energy electron or ion beams. We attribute the observed Lyman-α emission to the near-resonant energy transfer from the Ne_2^* excimer (14.5 – 15.5 eV corresponding to an emission wavelength of 80-85 nm) formed in the high-pressure discharge plasma to H_2 leading to the dissociation of H_2 (which requires 4.48 eV) and the excitation of the n=2 state of H (which requires 10.2 eV). Time-resolved fluorescence spectroscopic studies of both the excimer radiation and the Lyman-α emission have been carried out in an effort to elucidate the details of this near-resonant energy transfer process. The emission of the Lyman-β line radiation is attributed to a somewhat different near-resonant energy transfer process involving the first Ne excimer continuum and Ne metastable atoms.

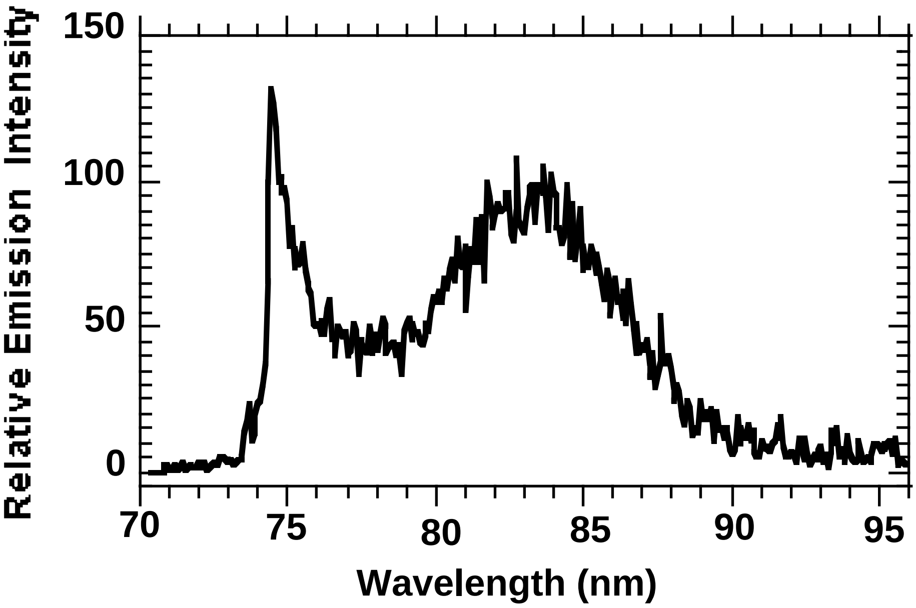

Fig. 2: Emission spectrum in the 70 –95 nm region from a MHC discharge in 740 Torr Ne.

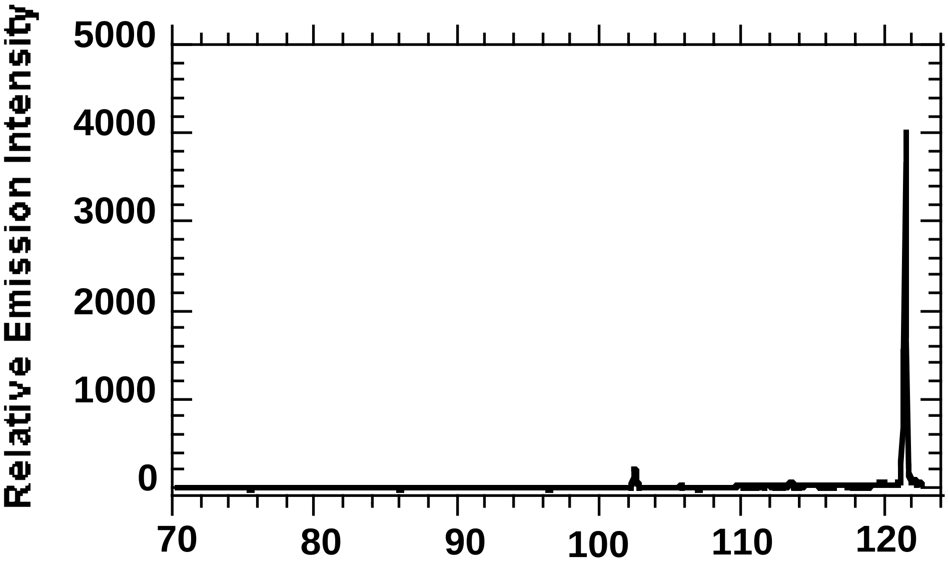

Fig. 3: Emission spectrum in the 70 – 125 nm region from a MHC discharge in 740 Torr Ne with a 1.5 Torr admixture of H_2.

4.2 Helium MHC Discharge Plasmas

Very recently [6], we have also observed (very intense) He_2^* excimer emission from a MHC discharge plasma operated in high pressure He (up to 600 Torr). Similar to what is shown in fig. 2 for Ne, the He excimer emission was found to consist of a narrowly peaked first He_2^* excimer continuum at 58 nm and a very broad second He_2^* excimer continumm from 60 nm to almost 100 nm. The presence of the He_2^* excimer emission is a clear indication that MHC discharge plasmas of the type studied here are very efficient sources of energetic electrons. For instance, the formation of the He_2^* excimer requires copious amounts of electrons with energies in excess of 20 eV.

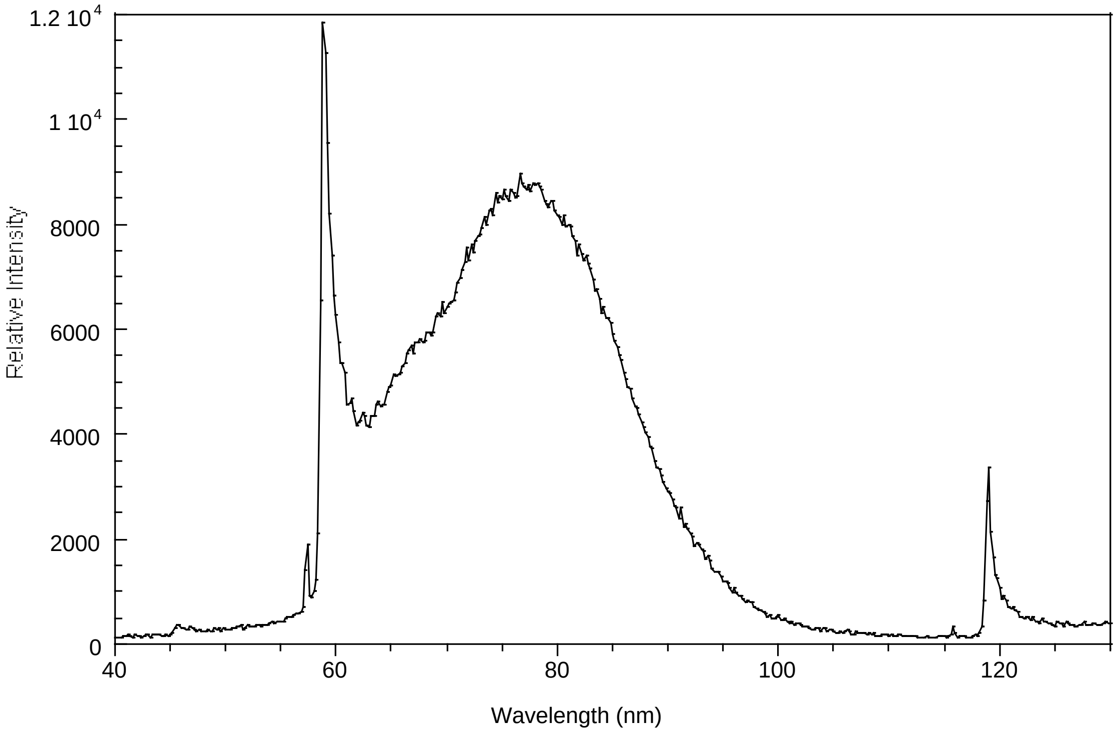

Fig. 4: Emission spectrum in the 60 –125 nm region from a MHC discharge in 400 Torr He. The emission feature around 120 nm represents the He excimer emission observed in second order.

Acknowledgments

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