1998

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Original Publication Citation

El-Habachi, A., & Schoenbach, K. H. (1998). Emission of excimer radiation from direct current, high-pressure hollow cathode discharges. *Applied Physics Letters, 72*(1), 22-24. doi:10.1063/1.120634

Emission of excimer radiation from direct current, high-pressure hollow cathode discharges

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(Received 15 July 1997; accepted for publication 4 November 1997)

A novel, nonequilibrium, high-pressure, direct current discharge, the microhollow cathode discharge, has been found to be an intense source of xenon and argon excimer radiation peaking at wavelengths of 170 and 130 nm, respectively. In argon discharges with a 100 μm diam hollow cathode, the intensity of the excimer radiation increased by a factor of 5 over the pressure range from 100 to 800 mbar. In xenon discharges, the intensity at 170 nm increased by two orders of magnitude when the pressure was raised from 250 mbar to 1 bar. Sustaining voltages were 200 V for argon and 400 V for xenon discharges, at current levels on the order of mA. The resistive current–voltage characteristics of the microdischarges indicate the possibility to form arrays for direct current, flat panel excimer lamps.
[S0003-6951(98)03501-3]

Excimer lamps are quasimonochromatic light sources that can be operated over a wide range of wavelengths in the ultraviolet (UV) and vacuum ultraviolet (VUV). The operation of excimer lamps is based on the formation of excited molecular complexes (excimers) and the transition from the bound excimer state to a repulsive ground state. In order to generate excimer radiation at high efficiency two conditions need to be satisfied:[1] the electron-energy distributions need to contain a sufficient concentration of electrons with energies required for the generation of the precursors of the excimers, which for rare-gas dimers are excited and ionized rare-gas atoms. Second, since the formation of the excimers is a three-body process, the pressure needs to be high, possibly on the order of one atmosphere or more. Both conditions can only be satisfied simultaneously in nonequilibrium discharges.

There are two ways to generate nonequilibrium plasmas: either by operating on a short enough time scale such that thermalization of the electrons is prevented, or by operating on a small enough spatial scale, e.g., in the cathode fall of a gas discharge. The first concept utilizes barrier discharges or silent discharges for excimer lamps.[1] The second type of nonequilibrium discharges are of such a size that they do not allow the development of a positive column, but only the electrode fall regions and the negative glow as, e.g., hollow cathode discharges.[2] The electrode geometry consists of a cathode, which contains some kind of a hollow and an arbitrarily shaped anode. The hole diameter, D, scales inversely with pressure, p, up to pressures of approximately 5–10 Torr/D(cm) for noble gases.[3,4] Atmospheric pressure operation in argon was, therefore, expected to be possible by reducing the hole diameter to values on the order of 100 μm and below.

The electrode geometry for the atmospheric pressure discharge is shown in Fig. 1, together with an end-on photograph of a microhollow cathode discharge in argon at 1.05 bar. The electrodes consist of molybdenum foils of 100 μm thickness, separated by a mica spacer of 200 μm thickness, and a cathode opening of 100 μm diam. Spectral measurements have been performed using a 0.5 m McPherson scanning monochromator, model 219, with a grating of 600 G/mm blazed at 150 nm. The discharge chamber with a MgF_2 window was mounted directly at the inlet of the monochromator. The spectrally resolved radiation at the exit slit was detected with a photomultiplier tube (Hamamatsu model R375) after conversion to visible light, centered around 425 nm, by a sodium salicylate scintillator. The entire system, including the monochromator, the space between the source and the entrance slit, and the space between the exit slit and scintillator was evacuated to a pressure of 60 μbar. The instrument resolution was ~2 nm full width at half maximum. The spectra were not corrected for detector response or for window transmittance.

We have concentrated on the VUV emission from microhollow cathode discharges in argon and xenon. Excimer emission from such discharges has been observed previously.[4,5] However, the discharges tended to become unstable at high currents and the emission changed from dc to pulsed.[4] By reducing the cathode hole diameter to 100 μm we were able to extend the current up to 7 mA (a value which was not exceeded because of concerns about thermal

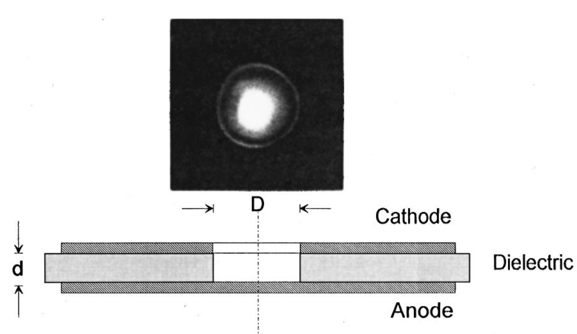

FIG. 1. Cross section of the hollow cathode geometry and an end-on photograph of a microhollow cathode discharge in argon at 1.05 bar, and a current of 1 mA. The circular cathode opening has a diameter of $D=100$ μm. The mica spacer has a thickness of 200 μm.

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 0003-6951/98/72(1)/22/3/$15.00

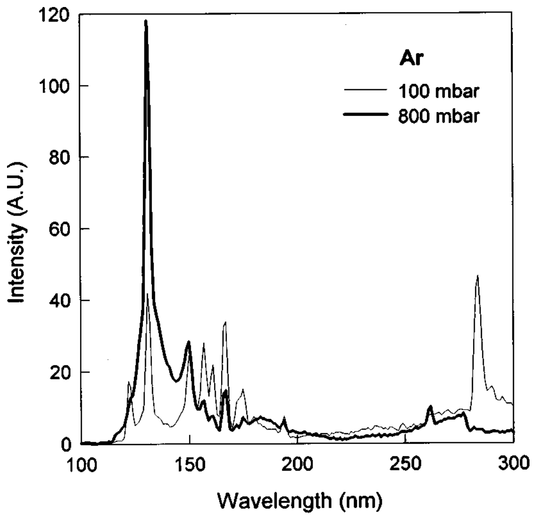

FIG. 2. VUV spectra of discharges in argon at 100 and 800 mbar. The sustaining voltage for the lower-pressure discharge was 221 V and the current was 5 mA. For the higher-pressure discharge the voltage was 200 V, and the current was 5.4 mA.

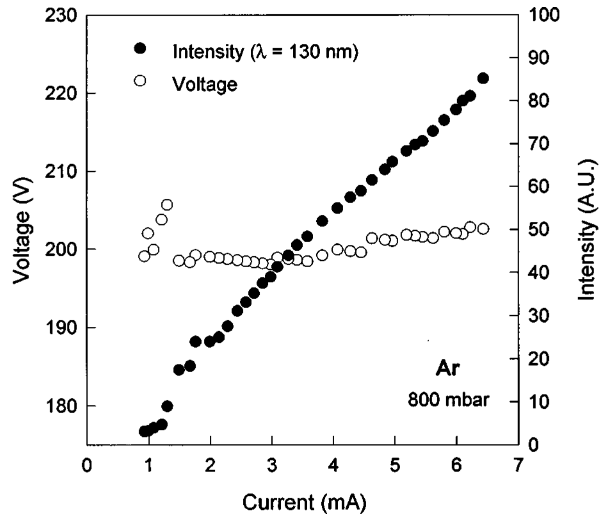

FIG. 3. Voltage–current characteristics of microhollow discharges in argon (800 mbar) and corresponding current-dependent intensities of the argon excimer radiation at 130 nm.

damage of the electrodes) and still operate the discharge in a dc mode for both argon and xenon. Measurements in neon and nitrogen in a discharge geometry similar to ours, but with hole diameters of 200–400 μm have shown strong emission in the UV range,[6] however, the limited spectral range of the optical system did not allow the observation of neon excimer radiation.

The argon spectrum for discharge operation with hollow cathodes of 100 μm diam at pressures of 100 and 800 mbar is shown in Fig. 2. At low pressure (100 mbar), the spectrum over the range of 100–200 nm is dominated by Ar II lines, mostly transitions between states having a $3s^2 3p^4(^3P)$ ionic core.[7] At high pressure the intensity of these lines is strongly reduced and the main spectral feature is the excimer line. The peak of the excimer line was observed at 130 nm. Due to a rapid change of the transmittance of the MgF_2 window between 125 and 130 nm (53% and 60%, respectively), and a similar reflectivity change of the MgF_2-coated mirrors inside the monochromator, the actual peak could be at slightly shorter wavelength. The argon discharge was operated in flowing gas, at 380 sccm. Operating the discharge without flow caused a reduction of the intensity by more than an order of magnitude. A possible reason for this decay is the increased contamination of the gas by electrode vapor.

The emission of the argon excimer radiation is, besides on the gas pressure, also dependent on the discharge current. The sustaining voltage, V, of the argon microhollow cathode discharge and the intensity of the excimer radiation is plotted versus the discharge current, I, in Fig. 3. Typical sustaining voltages are 200 V. The $I-V$ characteristic has a positive slope over most of the current range, similar to an abnormal glow discharge in a plane–plane geometry. This is expected for a hollow cathode discharge at high current.[8] In the current range of zero and positive differential resistivity the intensity of the excimer line, measured at 130 nm, increases linearly with current. Regions of negative differential resistivity, as the one seen in Fig. 3 at a current of 1.3 mA, are correlated with a nonlinear increase in excimer emission. This effect, which was found to be even more pronounced in xenon, however at higher current, is assumed to be caused by the transition from a glow discharge into a hollow cathode discharge with an increased rate of ionization due to pendulum electrons.[2,8] Another possible cause for the observed negative differential resistivity is the increased ionization of metastable states with increasing current. Such a multistage ionization process causes discharge instability, the so-called ionization instability,[9] which is associated with a negative voltage–current characteristic.[10] The increase in ionization causes a drastic increase in the concentration of molecular ions generated through three-body collisions of the atomic ions with atoms. This process in turn leads to an increased formation of excimers, with a consequent increase in the intensity of excimer radiation.

Experiments in xenon were only performed in a static gas environment. The spectra of xenon at pressures of 100 and 1000 mbar are shown in Fig. 4. Most of the lines seen in

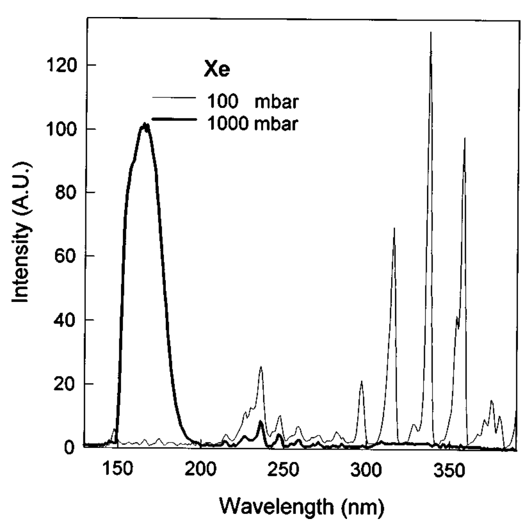

FIG. 4. VUV spectra of discharges in xenon at 100 and 1000 mbar. The sustaining voltage for the lower-pressure discharge was 427 V and the current was 4.75 mA. For the higher-pressure discharge the voltage was 395 V, and the current was 4.8 mA.

the xenon spectrum at low pressure are Xe II lines. The line at 315 nm was identified as a molybdenum (electrode material) line. It is also visible in the argon discharge spectrum. Its reduced intensity at higher pressure indicates less sputtering of the electrode material. At atmospheric pressure the main feature in the xenon spectrum is the excimer line with a measured maximum emission at 170 nm. The sustaining voltage for xenon discharges is twice as high as for argon discharges, 400 V for xenon versus 200 V for argon.

The observed excimer emission points to the presence of a large concentration of electrons with energies on the order of the metastable argon atom (12.5 eV). The Ar II lines seen at low pressure indicate even higher electron energies, on the order of 30 eV. This is not too surprising since electron-energy measurements in hollow cathode discharges at low pressure[11] and in the negative glow of low-pressure discharges[12] have shown the presence of electrons with energies even greater than 100 eV. Extending the discharge operation to high pressure by reducing the electrode dimensions seems to only reduce the density of electrons in the high-energy tail of the electron-energy distribution but not eliminate them.

High-pressure operation, however, favors the generation of excimers, a three-body process. This is obvious from the pressure dependence of the excimer emission. In xenon, radiation intensity at 170 nm increased by two orders of magnitude when the pressure was raised from 250 mbar to 1 bar. In argon, the intensity of the excimer radiation at 800 mbar was five times that at 100 mbar. This pressure dependence can be explained by considering the rates for the generation of excited rare-gas atoms R^*, the precursors for the excited rare-gas molecules R_2^*:

$$\frac{d[R^*]}{dt} = W' - kR^*R^2 - \frac{R^*}{\tau_a},$$

where k is the rate coefficient for three-body collision and W' is the source term for the generation of excited atoms. The apparent lifetime of the excited atoms, τ_a, is besides being gas dependent also determined by radiation trapping.[13,14] For efficient excimer formation, the three-body collision term kR^*R^2 needs to exceed the radiative decay term R^*/τ_a. The value of the onset pressure for efficient excimer radiation can, therefore, be estimated by setting the two terms equal. For xenon, k is 5×10^{-32} cm^6/s,[15] and τ_a, which scales with the square root of the plasma cell dimensions,[14] is estimated to be 1 μs in the microhollow cathode geometry. Using these values, the onset pressure for excimer radiation in xenon is 180 mbar. Since diffusion was neglected as an additional loss process, the onset pressure is actually higher. This is in agreement with our experimental results, where excimer radiation in xenon becomes observable above 250 mbar.

The fact that the current–voltage characteristic of microhollow discharges has a positive slope over a large range of current allows the operation of these discharges in parallel,[16] and consequently the construction of flat panel excimer lamps. A microhollow cathode discharge excimer lamp has the potential to operate dc, at voltages of several hundred volts. The simplicity of the electrode geometry, which can be generated by means of thin-film technology, may allow the formation of thin, almost two-dimensional excimer lamps, which can easily be adapted to any load geometry.

The authors like to thank U. Kogelschatz (ABB Corporate Research) for stimulating discussions. This work is supported by the U. S. Department of Energy, Advanced Energy Division.

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