2000

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Microhollow cathode discharge excimer lamps*

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(Received 15 November 1999; accepted 20 January 2000)

Microhollow cathode discharges are high-pressure, nonequilibrium gas discharges between a hollow cathode and a planar or hollow anode with electrode dimensions in the 100 μm range. The large concentration of high-energy electrons, in combination with the high-gas density favors excimer formation. Excimer emission was observed in xenon and argon, at wavelengths of 128 and 172 nm, respectively, and in argon fluoride and xenon chloride, at 193 and 308 nm. The radiant emittance of the excimer radiation was found to increase monotonically with pressure. However, due to the decrease in source size with pressure, the efficiency (ratio of excimer radiant power to input electrical power), has for xenon and argon fluoride a maximum at ~400 Torr. The maximum efficiency is between 6% and 9% for xenon, and ~2% for argon fluoride. © 2000 American Institute of Physics. [S1070-664X(00)95105-X]

I. INTRODUCTION

Excimer lamps are quasi-monochromatic light sources, which 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. Examples for these complexes are rare-gas dimers and rare-gas–halogen exiplexes. The advantage of excimer lamps over other spectral lamps is their high-internal efficiency, which may reach values of up to 40%, when operated at high pressure. The fact that it is a noncoherent radiation source allows us to scale the lamp to large size and to use it to irradiate (and treat) large areas. Applications for excimer lamps are UV curing and polymerization, UV oxidation, photo-chemistry, photo-deposition, photo-annealing, pollution control, to name only a few.2

In order to generate excimer radiation two conditions need to be satisfied: First, the electron energy distribution needs to contain a sufficient concentration of electrons with energies larger than the excitation energy of the excimer gas atoms. Secondly, since the formation of excimers is a three-body process, the pressure needs to be high, on the order of one atmosphere or higher. Both conditions can only be satisfied simultaneously in nonequilibrium plasmas. There are two ways to generate nonequilibrium plasmas: Operation at high-electric fields on such a short time scale that thermalization of the plasma is prevented, or operation on a small enough spatial scale, e.g., in the cathode fall of a gas discharge. The first concept is used in barrier (silent) discharges, discharges between dielectric covered electrodes separated by gas filled gaps of millimeter to centimeter distance. The second kind of nonequilibrium plasmas is found in plasma boundary layers, particularly the cathode fall of stable high-pressure discharges, such as corona discharges and high-pressure hollow cathode discharges. We have studied the latter type, hollow cathode discharges, with respect to the application as excimer source.

Hollow cathode discharges are gas discharges between a cathode, which contains a hollow structure, and an arbitrarily shaped anode. At gas pressures such that the pressure, p, times cathode hole diameter, D, is on the order of Torr cm, the discharge develops in stages, dependent on the discharge current. At low currents a "predischarge" is observed, a glow discharge with a shape determined by the vacuum electric field. With increasing current, the plasma column formed along the axis of the cathode hole begins to serve as a virtual anode, causing a modification of the electric field distribution in the cathode hole. The initially axial electric field in the cathode plane changes into a radial one, and electrons, generated at the cathode, are accelerated radially towards the axis. They lose their energy in the cathode fall and in the negative glow, which for high values of pD is a ring shaped plasma layer adjacent to the cathode edge. For small values of pD, on the order of 1 Torr cm and less, the negative glow extends to the center forming a plasma cylinder.

When the discharge changes from an axial predischarge into a radial discharge the sustaining voltage drops and the current increases: The discharge has a negative differential resistance. Although discharges in all current modes are hollow cathode discharges, generally the term "hollow cathode discharge" is used only for this mode. With increasing current the discharge voltage stays first constant, typical for a normal glow discharge, and then transfers into an abnormal glow discharge, characterized by a positive differential resistance.

Hollow cathode discharges are known for an electron energy distribution, which contains a high concentration of high-energy electrons. Using spectral diagnostics,7 retarding
field analyzers, and probes electron energies well over 10 eV have been measured. But most of the studies have been performed in low-pressure hollow cathode discharges. Attempts to extend the range of pressure to higher values have been reported by White in 1958. According to the White–Allis similarity law, \( V = V(pD, I/D) \), where \( V \) is the sustaining voltage, and \( I \) is the discharge current, higher-pressure operation can be achieved by reducing the diameter, \( D \), of the cathode hole. The lowest value of \( pD \), for which this law holds, is given by the condition that the mean free path for ionization must not exceed the hole diameter. For argon, the minimum \( pD \) is, according to this condition, 0.026 Torr cm. Empirical values for the upper limit in \( pD \) are 10 Torr cm for rare gases, less for molecular gases. Based on the assumption, that electrons oscillating through the center between opposite cathode falls (pendulum electrons) are responsible for the “hollow cathode effect,” the upper limit for \( pD \) can be determined by the condition that the distance between opposite cathodes must not exceed the lengths of the two cathode fall lengths plus the negative glow. This leads for argon to an upper limit in \( pD \) of slightly more than 1 Torr cm.

Because of the required small size of the cathode opening for high-pressure operation we have coined the term “MicroHollow Cathode Discharges (MHCD)” for these discharges. For atmospheric pressure discharges typical hole diameters should, according to the upper limit value for \( pD \), be on the order of ten micrometers. However, this value is based on the assumption that the gas is at room temperature, a condition, which is not fulfilled in MHCDs. In these discharges the gas temperature may reach values of 2000 K, as shown for atmospheric pressure MHCDs in air. Since the gas density, rather than the gas pressure is the relevant parameter in the similarity law for hollow cathode discharges, the temperature needs to be taken into account. If the effect of three-body collisions is neglected and the ideal gas law is applied the similarity law can be corrected for temperature dependence by multiplying the \( pD \) value with the ratio of actual gas temperature to room temperature. But even taking the relatively high-gas temperature of MHCDs into consideration, the diameter of the cathode opening should still be less than 100 \( \mu \)m for hollow cathode discharge operation. However, stable hollow cathode discharges have been observed with cathode hole sizes as large as 250 \( \mu \)m in xenon. These results indicate that at high \( pD \) values photon coupling rather than pendulum–electron coupling between opposite cathode falls is responsible for the observed negative differential resistance and the discharge stability.

II. EXPERIMENTS

The electrode geometry for a single hole microhollow cathode excimer lamp as it is used in our experiments is shown in Fig. 1. The electrode geometry consists of two metal plates with circular opening, separated by a dielectric film. This geometry is a simplified version of hollow cathode discharge geometries where the cathode contains a cylindrical hole or a cylindrical cavity. Both, cathode and anode consist of 100 \( \mu \)m thick molybdenum. In earlier experiments they were separated by a mica layer of 200 \( \mu \)m. In more recent experiments we have used 100–250 \( \mu \)m thick alumina \( (\text{Al}_2\text{O}_3) \) because it withstands higher temperatures. The cylindrical holes in the cathode and the mica have been varied between 80 and 700 \( \mu \)m.

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, and by means of a 0.2 m McPherson monochromator, model 234/302, with a grating of 1200 G/mm. The discharge chamber with MgF\(_2\) or LiF\(_2\) windows was mounted directly at the inlet of the monochromator. The spectrally resolved radiation at the exit slit was detected with a photomultiplier tube after conversion to visible light, centered around 425 nm, by a sodium salicylate scintillator. With slits opening of 600 \( \mu \)m the instrument resolution was ~3 nm full width at half maximum (FWHM).

In addition to spectral measurements we have measured the spatial distribution of the excimer source by using a VUV imaging system which allows us to generate an image of the excimer source with a magnification of ten onto the cathode of a proximity focused image converter. The emission from the fluorescent anode of the image converter is recorded by means of a charge coupled device (CCD) camera. Spectral resolution is obtained by using filters, which only allow the excimer radiation to pass.

III. RESULTS

A. Electrical characteristic and shape of excimer source

The dc (direct current) voltage characteristics of microhollow cathode discharges in rare gases show a distribution typical for hollow cathode discharges even for \( pD \) values large compared to 1 Torr cm. A current-voltage characteristic for discharges in xenon at a pressure of 750 Torr is shown in Fig. 2(b). The hole diameter is 250 \( \mu \)m, \( pD \) is consequently 18.75 Torr cm. For low current the differential resistivity of the discharge is positive, as expected for a hollow cathode discharge in the predischarge phase, where space charge effects (virtual anode) are not important. At a current of 4 mA the discharge enters a range with negative differential resistivity, the phase where it changes into a hollow cathode discharge with radial electric fields.

The source of the xenon excimer radiation, the microhollow cathode discharge plasma, as seen end-on through a band pass filter (maximum transmission of 24.4% at 170.9 nm and a FWHM of 26.8 nm) is only at low currents concentrated in the cathode hole [Fig. 2(a)]. The ring shaped region at the inner edge of the cathode opening represents the negative glow of the discharge. With increasing current, the excimer source extends into the area outside the hole,
covering at a current of 7 mA and a pressure of 750 Torr the cathode surface over a distance of approximately four times the hole diameter. As with current, the size of the excimer source changes with pressure. At high pressure, as for small currents, the source is located in or close to the cathode opening, particularly at the inner edge of the cathode hole (Fig. 3). With reduced pressure the source extends more and more over the cathode surface.

B. Spectral emission

Most of our excimer studies have focused on xenon, with its excimer emission peaking at 172 nm. A xenon spectrum for discharge operation with hollow electrodes of 100 μm diameter is shown in Fig. 4.18 At 40 Torr, the 147 nm xenon resonance line, corresponding to transitions from the $^3P_1$ state to the $^1S_0$ ground state, dominates the emission spectra. There are some indications of the first continuum, which extends from the resonance line towards longer wave-length. The second excimer continuum peaking at 172 nm appears at higher pressures. At pressures greater than 300 Torr, it dominates the emission spectra up to the longest recorded wavelength of 800 nm. The second continuum results from transitions from the lowest vibrational level of singlet $^1\Sigma$ and the triplet $^3\Sigma$ excimer states to the repulsive ground state. Transitions from higher vibrational levels of these states correspond to the first excimer continuum. Besides of pressure, the excimer emission is dependent on the discharge current. As shown in Fig. 2(b) it increases linearly with current above the transition into the hollow cathode mode.

Argon excimer emission has been studied in flowing gas with a gas flow of 380 sccm.19 At low pressure, the spectrum over the range of 100–200 nm is dominated by Ar II lines, mostly transitions between states having a $3s^23p^n$ ($^3P$) ionic core. At high pressure the intensity of these lines is strongly reduced and the main spectral feature is the excimer line, peaking at 130 nm. The emission of the argon excimer radiation increases, as for xenon, with gas pressure, and with discharge current. The sustaining voltage, $V$, is approximately the same as for xenon (200 V).

The presence of an attaching gas in the gas mixture is considered a major obstacle for the generation of high-pressure dc glow discharges. High-pressure discharges in rare-gas–halide mixtures tend to constrict and become unstable in times on the order of ten nanoseconds. However, as in rare gases, microhollow cathode discharges could also be operated in rare-gas–halogen mixtures in a stable dc mode up to atmospheric pressure. Argon fluoride excimer emission with a maximum at 193 nm was recorded in a gas mixture consisting of 1% fluorine, 5% argon, and 94% helium.20 The measured ArF excimer spectrum is shown in Fig. 5 for a pressure of 400 Torr. The line width at half intensity is just 3 nm, compared to 24 nm for Xe (Fig. 4). Similarly, xenon
chloride excimer radiation was measured peaking at 308 nm, in a gas mixture consisting of 0.06% hydrogen chloride, 0.03% hydrogen, 1.5% xenon and neon as buffer gas. Discharge voltages were ~500 V for discharges in ArF, and 180 V for XeCl discharges.

C. Efficiency and radiant emittance

Measurements of the efficiency, the ratio of radiant power in the UV to input electrical power, have been performed for xenon and argon fluoride discharges. In order to determine the absolute values of the excimer emission two methods have been used.\(^1\) One is based on comparing the discharge emission with that of calibrated UV sources: A Mercury vapor lamp (line emission at 185 nm) and a Deuterium lamp (continuum from 160 to 400 nm). A second one utilizes a calibrated radiometer. Both, for xenon and argon fluoride discharges, the efficiency was found to increase with pressure up to ~400 Torr, where it reaches values of 6%–9% for xenon\(^1\) and ~2% for ArF, and then decreases again for higher pressure. Although the efficiency for ArF is less than that for Xe, the peak spectral radiant power at identical electrical power input is for ArF higher by a factor of 2 to 3 compared to Xe, due to the differences in line width (Figs. 4 and 5).

The decrease of the measured efficiency at higher pressure can be explained by the decrease in size of the excimer source with pressure (Fig. 3). The radiant emittance, the optical power emitted per surface element, increases with pressure. However, this increase does, at pressures greater than 400 Torr, not compensate for the reduction in size of the source. Consequently, the overall optical power (integral of radiant emittance over source area) decreases at higher pressures.

D. Direct current versus pulsed operation

One of the special features of microhollow cathode excimer sources is their stability, which allows us to operate them in a direct current mode. However, in certain cases it could be an advantage to operate the discharges in a pulsed mode. This is particularly the case when high-radiant emittance is required. As known from dc measurements, the total optical power increases with current, however, at constant pressure the source size also increases [Fig. 2(a)]. The source area shrinks when the pressure is increased (Fig. 3). High-radiant emittance, therefore, requires both, high current and high pressure. The current for dc operation is limited by the thermal loading of the electrode structure to ~10 mA per discharge. With pulsed operation thermal loading of the electrodes can be largely avoided. The limitation in current, and consequently in intensity, is for pulsed operation determined by the development of current driven instabilities, the glow-to-arc transition, rather than thermal processes affecting the electrodes.

We have studied the discharge in xenon under pulsed condition and were able to extend the current range to 80 mA before instabilities set in.\(^2\) The temporal development of current, voltage, and excimer intensity of a discharge in xenon at a pressure of 300 Torr is shown in Fig. 6. Breakdown occurs at voltages between 700 V and 1 kV. A stable discharge phase is reached after times on the order of 100 \(\mu s\), dependent on discharge current.\(^2\)

E. Parallel operation

Industrial applications of microhollow cathode discharge excimer lamps require generally higher total optical power levels than achievable with single microhollow cathode discharges. The optical power of single xenon discharge reaches approximately hundred mW (at an efficiency of 6%–9%), consequently, the operation at the kW optical power level would require an array of more than \(10^4\) discharges.

It can be expected that in the current range where the \(V-I\) characteristic of the microhollow cathode discharge has a positive slope, the Townsend region and the abnormal glow region,\(^5\) parallel operation of microhollow cathode discharges can be achieved without ballasting the individual discharges. Operation of the discharge in the abnormal glow mode requires limiting of the cathode area, such that the
current density increases with increasing current. One way to limit the cathode area is to use blind holes in the cathode material instead of openings as shown in Fig. 1. Such geometry was utilized in earlier experiments, and parallel operation could be demonstrated. A second method is to use a geometry as shown in Fig. 1, but to cover the cathode area with a dielectric, except the cylindrical surface of the cathode opening. This method has allowed us to operate two microhollow cathode discharges in atmospheric pressure argon in parallel without individual ballast.

For discharges operating in modes where the slope of the current-voltage characteristics is negative, flat, or only slightly positive, it was not possible to obtain stable parallel dc operation consistently without ballasting the individual discharges. Individual ballasting is a reasonable approach for relatively small arrays. A method, which has allowed us to extend this method to large arrays, is the use of distributed resistive ballast. This was achieved by using a semi-insulating material, in our case semi-insulating silicon, as anode material. The result of this experiment is shown in Fig. 7. The method allows us to generate arrays of microhollow cathode discharge excimer sources of any size, limited only by the thermal loading of the ballast resistor.

IV. DISCUSSION

One important condition for excimer formation in glow discharges is an electron energy distribution with a large concentration of electrons with energies greater than the excitation energy of the excimer gases. For argon, the lowest excited state is at 11.55 eV. Excimer emission using the same microhollow cathode geometry as shown in Fig. 1 has even been reported for neon, where the lowest excited state is at 16.6 eV. The presence of electrons in hollow cathode discharges with energies greater than the energy required for populating the lowest excited states in rare gases is not surprising. It has been shown in various experiments that the electron energy distribution in such discharges contains a high concentration of electrons in the high-energy tail of the distribution (for example, see Ref. 4). Measurements of the energy distribution of electrons accelerated in the cathode fall of a glow discharge have shown that even a beam component exists with electron energies comparable to the full cathode fall energy.

The second condition for dc excimer sources, a stable, nonthermal discharge at high-neutral gas density (such that excimer formation, a three-body process, occurs at a higher rate than collisional or radiative decay of the excimer precursors) can, to our knowledge, only be fulfilled in corona discharges and micro discharges, such as the MHCD. High-pressure glow discharges in plane parallel electrode geometries are prone to instabilities, particularly to glow-to-arc transitions, and can only be sustained for times on the order of ten nanoseconds.

The exceptional stability of hollow cathode discharges is assumed to be due to a coupling process between the two cathode falls, which face each other at a distance of D. For low pD values, it is the electrons which provide coupling between opposite cathode sheaths. The “pendulum” electrons generated at the cathode surface and accelerated in the cathode fall, gain enough energy to oscillate through the plasma on axis, which serves as a virtual anode. Whenever instabilities, characterized by locally increasing electron density, begin to form in the hollow cathode fall, the voltage across the cylindrical cathode fall is reduced. This causes a reduction in the concentration of pendulum electrons and consequently a reduced ionization rate in opposite cathode falls. This effect counters the growth of electron density, and would explain the excellent stability of hollow cathode discharges.

However, for values of pD exceeding 1 Torr cm (at room temperature), electron coupling between the opposite cathode falls becomes increasingly unlikely. Still, hollow cathode discharges in xenon were shown to have a hollow cathode discharge phase with negative differential resistance even for temperature corrected pD values large compared to one Torr cm. At high values of pD, it is assumed that photons provide the coupling between opposite cathode falls. Photoelectron emission from the cathode surfaces has been suggested by Little and von Engel as a mechanism responsible for the hollow cathode effect. Experiments in argon between two parallel cathodes of variable distance seem to support this assumption. An effect on discharge current and sustaining voltage was observed at “hollow” cathode distances large compared to the distance where the two negative glows merged. Merging occurred at a pD value close to 1 Torr cm, in agreement with computational results. As in the case of electron coupling, the reduction in photoemission, caused by reduced cathode fall voltage due to the emergence of a local increase in electron density (instability) counters the growth of this instability.

The high energies of electrons accelerated in the cathode fall, and the excellent stability of microhollow cathode discharges are the important features for microhollow cathode discharge excimer lamps. An additional bonus is the fact that by increasing the current (abnormal glow) or by decreasing it (predischarge) the discharge behaves like a resistor: A feature, which allows us to generate arrays of such discharges.
with relatively simple means. Experiments in xenon indicate that in MHCD arrays an average radiant emittance of 50 W/cm² may be achievable.

The measured efficiency of 6%–9% for xenon excimer MHCD emitters is a factor of five below theoretical values. The reason for the lower than expected efficiency is assumed to be the relatively high temperature of the plasma. Heating an excimer gas is known to reduce the excimer emission. This is due to the reduction in gas density with increasing temperature at a fixed pressure, but also due to the fact that the rate coefficient of the three-body process, which leads to excimer formation, scales with $T^{0.5}$. Temperature measurements in microhollow cathode discharges have to date only been performed in atmospheric air, where temperatures of ~2000 K have been obtained. Although the gas temperature in rare-gas microhollow cathode discharges is expected to be lower, it is probably still large enough to cause a strong reduction in efficiency. Cooling through gas flow is expected to increase the efficiency of MHCD excimer sources.

Barrier discharge excimer lamps have, probably to a large extend due to the lower plasma temperature, a higher efficiency than MHCDs. Whereas the highest measured internal efficiency in MHCDs is 9% for Xe, corresponding to an external efficiency of approximately half this value, external efficiencies of 10% have been measured in barrier discharge lamps, and even 20% seemed to be achievable. However, the radiant emittance, which is stated in Ref. 31 as being in the range of 100 mW/cm² is considerably higher for MHCDs where an emittance of 10 W/cm² has been reached.

A particular feature of microhollow cathode discharges is the extreme dc power density in these discharge plasmas. Typical discharge voltages are ~200 V. Currents of up to 10 mA are reached in such discharges when operated dc, resulting in an electrical power for single discharges on the order of Watt. For high-pressure operation the plasma is concentrated in the cathode opening. The volume of cathode fall and negative glow, where most of the electrical energy is dissipated is assumed to be less than the volume determined by the cathode opening. For a 100 μm thick, 100 μm diam cathode opening the volume is 0.810^{-6} cm³. The power density is consequently on the order of 10⁶ W/cm³. In the pulsed mode, where we are able to reach 80 mA for millisecond time duration, the power density is expected to be even higher. With power densities that high conditions for lasing should be achievable when a multitude of such discharges is arranged in series. This opens the possibility to build cw (continuous wave) or quasi cw excimer microlasers, an exciting prospect for the future of microhollow cathode discharges.

ACKNOWLEDGMENTS

This work is supported by the Department of Energy, the National Science Foundation, the Defense Advanced Research Projects Agency, and the Air Force Office of Scientific Research. The authors would also like to thank Dr. Jaeyoung Park, Los Alamos National Laboratory, for helpful discussions.