Microhollow cathode discharges

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By reducing the dimensions of hollow cathodes into the hundred micrometer range, stable, direct current, high (atmospheric) pressure glow discharges in rare gases, rare gas–halide mixtures and in air could be generated. The electron energy distribution in these microdischarges is non-Maxwellian, with a pronounced high-energy tail. The high electron energy together with the high gas density, which favors three-body collisions, is the reason for an efficient excimer generation in these microplasmas. Excimer efficiencies from 1% to 9% have been measured for argon, xenon, argon fluoride, and xenon chloride direct current excimer emitters, with a radiant excimer emittance of up to 2 W/cm^2 for xenon. Adding small amounts of oxygen to argon has allowed us to generate vacuum ultraviolet line radiation at 130.5 nm with an efficiency approaching 1%. Pulsing xenon discharges with nanosecond electrical pulses has led to an increase in intensity to 15 W/cm^2 and to a simultaneous increase in efficiency to more than 20%. Operating the discharges in an abnormal glow mode has allowed us to generate microdischarge arrays without individual ballast. Applications of these plasma arrays are excimer lamps and plasma reactors.
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I. INTRODUCTION

Hollow cathode discharges are gas discharges between a cathode, which contains a hollow structure, and an arbitrarily shaped anode.[1] A typical hollow cathode structure would consist of a cylindrical hole in a cathode, with a ring shaped anode, separated by dielectric spacer [Fig. 1(a)],[2] or the anode could be just a metal pin.[3] The cathode ''hollow'' does not need to be a hole in a solid cathode; a cylindrical opening in a thin cathode layer, such as shown in Fig. 1(b) qualifies also as a hollow cathode structure.[4] The anode in this case is an electrode of similar shape [Fig. 1(b)] as the cathode, but it could also consist of a metal foil without hole.[4]

Modeling results[5] and experimental observations[4] show for discharges in such electrode geometries at gas pressures p and cathode hole diameters D, such that the product of these two parameters is on the order of 1 Torr cm, the existence of various discharge modes dependent on current. For very low currents, the discharge extends from the cathode surface through the cathode hole to the anode with the electric field in the cathode hole being axial. With increasing current, the plasma column formed along the axis of the cathode hole causes 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. For pressure values such that the diameter of the cathode opening is less than twice the length of the cathode fall region (cathode fall+negative glow) the electrons accelerated in the cathode fall reach the opposite cathode fall, where they are again accelerated and oscillate with ever decreasing amplitude between the opposite cathode falls. The increased ionization rate of such ''pendulum'' electrons[6] in the hollow cathode causes a decrease in voltage with increasing current (negative differential resistance).[5]

With a further increase of current the cathode layer expands over the surface of the planar cathode outside the hole. The current–voltage characteristic becomes that of a normal glow discharge with constant voltage at increasing current. Ultimately, when the cathode layer reaches the boundaries of the cathode, any further current increase requires an increase in discharge voltage: the discharge changes into an abnormal glow discharge. Although all discharge modes can be observed in hollow cathode configurations, the term ''hollow cathode discharge'' is generally used only for the discharge mode characterized by a negative differential resistance.

Based on the hypothesis that the hollow cathode effect is caused by ''pendulum'' electrons, hollow cathode discharges are expected to follow a similarity law:[3,7] $V=\mathrm{V(pD)}$, with V being the sustaining voltage for the hollow cathode discharge. The lowest value of pD is given by the condition that the mean free path for ionization must not exceed the hole diameter.[8] The upper limit for pD is determined by the condition that the distance between opposite cathodes must not exceed the lengths of the two cathode fall regions. This condition leads to values of slightly higher than 1 Torr cm as an upper limit for pD in argon.[4] Empirical values for this upper limit are even higher: 10 Torr cm for rare gases, less for molecular gases.[9] This discrepancy may be due to the assumption of the maximum distance between opposite electrodes for the ''pendulum'' electron effect to occur.[4] Experimental studies where the current-voltage characteristics of a hollow cathode discharge have been measured versus the distance between opposite cathodes have shown that the hollow cathode effect already sets in before the negative glows merge.[10] This increased interaction range shifts the calculated pD value[4] closer to the empirical one.[9]

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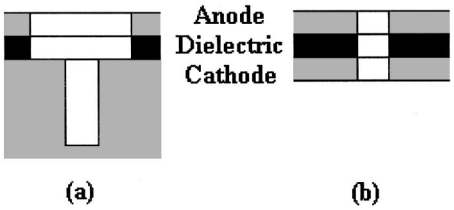

FIG. 1. Electrode geometries for microhollow cathode discharges.

The similarity law for hollow cathode discharges has been the basis of efforts to extend the pressure range for hollow cathode discharge operation to atmospheric pressures. By using cathodes with holes in the 100 μm diam range, it was possible to operate hollow cathode discharges at atmospheric pressure in rare gases,[4,11] in rare gas-halide mixtures,[12] and in air.[13] 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.[2]

II. MICROHOLLOW CATHODE DISCHARGE PARAMETERS

Both electrode geometries depicted in Fig. 1 have been used in microhollow cathode studies by us and by other research teams.[14–18] However, most of our studies have been performed with an electrode geometry as shown in Fig. 1(b). The reason for this choice is the simplicity of this electrode configuration, which allows easy manufacturing of large area micro electrode arrays by means of plasma spraying and laser drilling,[19] and the ability to flow gases through the discharge volume inside the hollow. Flowing allows the use of these discharges as plasma reactors, for thermal management (cooling of electrodes), and for the replenishment of gas contaminated by sputtered or evaporated electrode material. The use of a third, positively biased electrode placed at the anode side of microhollow cathode discharges, has allowed us to utilize the microhollow cathode discharge as an electron emitter and as a plasma cathode, respectively, for glow discharges between the MHCD and the third electrode.[20] Stable glow discharges with dimensions of up to centimeters, even in atmospheric pressure air, could be generated with this method.[21]

The electrodes used for plasma cathodes and for excimer source studies described in the following, consist of two molybdenum foils with circular openings separated by a dielectric film. In earlier experiments the electrodes were separated by a mica layer of 200 μm. In more recent experiments we have used 100–250 μm thick alumina (Al_2O_3) because it withstands higher temperatures. The cylindrical holes in the cathode and the mica have been varied between 80 and 700 μm. Holes with a diameter of less than 150 μm were drilled by means of an excimer laser, larger holes can be drilled using mechanical tools. The discharges were either operated dc or pulsed, with pulse durations either in the ms range or in the nanosecond range. The ms pulses with duty cycles of 0.007 allow us to increase the peak power without increasing the average power, and consequently extend the power range without causing thermal damage to the sample. Nanosecond pulsing provides a means to increase the electron energy with negligible gas heating.[22]

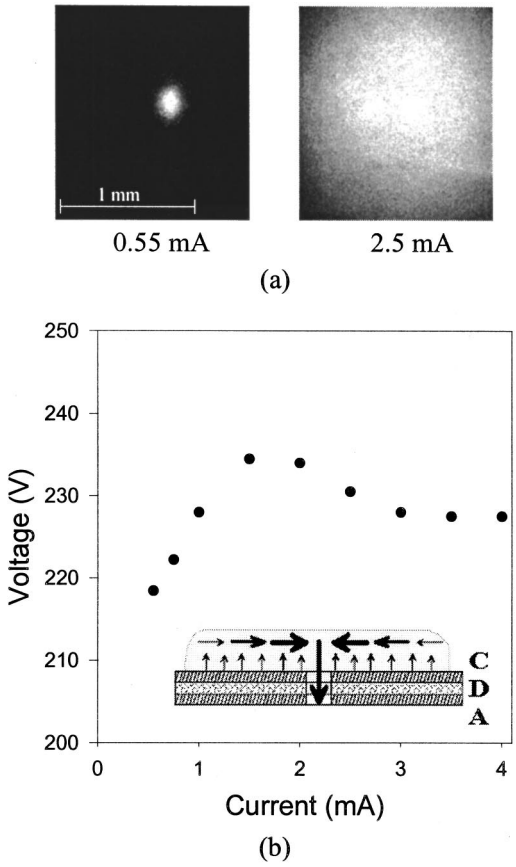

FIG. 2. (a) End-on view of microhollow cathode discharges in argon at pressures of 760 Torr for various discharge currents at wavelengths around 128 nm. The cathode hole has a diameter of 130 μm. The photograph on the left shows the discharge in the predischarge mode, the one on the right the discharge in the normal glow mode. The bright spot off center in the photograph on the right is due to am imperfection at the cathode surface. (b) Sustaining discharge voltage vs discharge current. The inset shows the cross section of the electrodes, and the current pattern in the normal glow discharge mode.

Diagnostic studies of microhollow cathode discharges include measurements of the current–voltage characteristics, temporally resolved photography of the discharges in both the visible and the ultraviolet (UV), measurements of the electron density (in argon discharges), measurement of the gas temperature (in air), and observations of the emission spectrum, mainly in the UV and vacuum UV (VUV). The spectral measurements in the UV and VUV, which were performed for rare gases and rare gas-halide mixtures, have focused on the spectral range of excimer emission.

The voltage and the appearance of microhollow cathode discharges changes with increasing current as described in the introduction. For argon, end-on VUV photographs [Fig. 2(a)] and the voltage–current diagram [Fig. 2(b)] show the development of the discharges in the predischarge mode, the hollow cathode discharge mode, and the normal glow discharge mode. The cross section of the electrodes and a draw-

ing, which depicts the current pattern for the normal glow mode, are shown in the insert in Fig. 2(b). For low current, the plasma is located in the center of the hole [Fig. 2(a), 0.55 mA]. It fills the hollow when the discharge is operated in the hollow cathode discharge mode (negative differential conductance), and for higher currents, expands into the area outside the hole [Fig. 2(a), 2.5 mA]. When it reaches this state, voltage and current density stay constant with increasing current, which is characteristic for normal glow discharges.

Electron density measurements in microhollow cathode discharges in argon with small admixtures of hydrogen have been performed utilizing Stark broadening of the hydrogen Balmer-β line at 486.1 nm.[23] For argon pressures of 760 Torr with 1% hydrogen added, electron densities of $10^{15}\,\mathrm{cm}^{-3}$ were measured for currents of 3 mA. The electron density increases linearly with current to values of $1.4\times10^{15}\,\mathrm{cm}^{-3}$ at a current of 10 mA. With 10 ns pulses of 600 V amplitude applied to the electrodes, peak electron densities of $5\times10^{16}\,\mathrm{cm}^{-3}$ were measured. This value is at least one order of magnitude higher than that obtained for any other high-pressure, nonthermal glow discharge.[24]

The gas temperature in microhollow cathode discharges has been measured for atmospheric air as working gas by evaluating the rotational (0–0) band of the second positive system of nitrogen.[25] For air, the temperature in the microhollow varies between 1700 and 2000 K at discharge currents between 4 and 12 mA. The temperature in rare gas microhollow cathode discharges has not yet been measured, but it is assumed that it is considerably less than the temperature in atmospheric pressure air discharges.

III. MICROHOLLOW CATHODE DISCHARGE ULTRAVIOLET LIGHT SOURCES

Hollow cathode discharges are known for an electron energy distribution which contains a high concentration of high-energy electrons. Using spectral diagnostics,[26] retarding field analyzers,[27] and probes,[28] electron energies well over 10 eV have been measured. Although all these measurements have been performed at low pressures, similar electron energy distributions can be expected at high pressures. The high-energy tail in the electron energy distribution is responsible for the measured high density of electrons[23] and for the high number density of excited atoms.

In rare gas and rare gas-halide discharges, excited rare gas atoms serve as precursors for excimers. The high-pressure operation favors three-body processes, such as excimer formation. It can therefore be expected that high-pressure microdischarges, operated dc or pulsed, will serve as strong sources of excimer radiation. For xenon, one of the most thoroughly studied excimer gases, the reaction equation is

$$\mathrm{Xe}^{*}+\mathrm{Xe}+\mathrm{Xe}\rightarrow\mathrm{Xe}_2^{*}+\mathrm{Xe}$$

with a rate constant of $k=5\times10^{-32}\,\mathrm{cm}^6/\mathrm{s}$ at room temperature.[29] The excited dimers decay radiatively into a repulsive ground state, which means that reabsorption processes are negligible; the plasma and excimer gas is optically thin for excimer radiation.

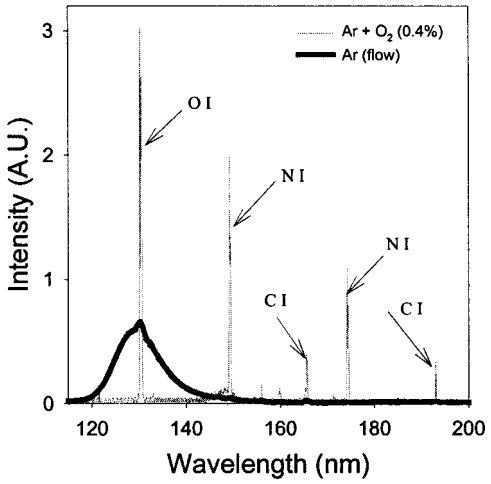

FIG. 3. Argon excimer spectrum for flowing gas operation (760 Torr), and a line emission spectrum obtained by adding 0.4% oxygen to 1100 Torr argon.

Previous results, mainly with xenon as excimer gas, operated dc, have been discussed in a review article.[12] Here we focus on some more recent results, particularly the effect of impurities on the excimer emission and results of pulsed discharge operation. It is known that even small concentrations of impurities are detrimental to excimer sources. It is therefore important to use only highly purified gases, evacuate the discharge chamber thoroughly before operation, and use only materials for the discharge chamber with low gas emission. Even small leaks and/or sputtered or evaporated electrode material will limit the lifetime of an excimer source considerably. The $1/e$ decay time of excimer emission in static xenon was, e.g., measured as 580 min.[30] A way to overcome the contamination problem to a large extent is to work with flowing gas. Whereas the VUV spectrum in static argon is dominated by impurity radiation, particularly atomic oxygen radiation at 130.5 nm, by flowing the gas, the impurity radiation could be largely suppressed.

On the other hand, impurities such as oxygen have a place as line emission sources for radiation in the VUV. Small additions of oxygen to argon at high pressure (1100 Torr) have allowed us to increase the oxygen line emission at 130.5 nm to values of 13 mW, at the expense of the argon excimer emission.[31] The argon excimer emission, without oxygen added, was measured as 16 mW. The spectrum of an argon discharge with 0.4% oxygen added is shown in Fig. 3 compared to an argon excimer spectrum in flowing argon. This strong line emission, which is assumed to be due to resonant energy transfer from argon dimers or argon dimer precursors to atomic oxygen, might have applications for VUV photolithography. Similar resonant energy transfer from neon dimers to atomic hydrogen (H Lyman-α line emission at 121.6 nm) has been observed by Becker *et al.*[32]

The excimer efficiency, defined as the ratio of optical power in the spectral range of the excimer emission to the

TABLE I. Parameters of microhollow cathode discharge ultraviolet sources.

Excimer	Pressure (Torr)	Voltage (V)	Current (mA) dc	Current (mA) ms pulses	Wavelength (nm)	Efficiency (%) dc/ms pulses	Efficiency (%) ns pulses	Ref.
Xe_2^*	≤760	220	1–8	<70	172	6–9	20	4,12,33, 34,37
Ar_2^*	≤1100	200	1–10	—	128	6	3–5	4,23,34
Ar_2^*(Ar+1% O_2)	≤1100	300	1–10	—	130.5	<1	—	31
XeCl*	≤1000	190	1–8	<200	308	3	—	38
ArF*	≤760	200	1–8	—	193	1	—	12

electrical power, was measured as 6% for flowing argon. The measurement was performed with a calibrated photodiode detector (SXUV-100 supplied by IRD), and isotropic optical emission was assumed. With applied voltages of 200 V and currents of 3 mA at the transition between hollow cathode discharge and normal glow discharge phase, the total excimer emission is 35 mW. Because of the assumed isotropic emission, only half of it reaches the observer. This value is slightly less than that obtained for xenon, which was measured as 6%–9% for dc operation,[33] but higher than obtained for ArF and XeCl excimer radiation, which was found to be 1% and 3%, respectively.[12] For all the gases, the excimer intensity was found to increase linearly with current at constant voltage (normal glow) and constant pressure.[34] The intensity (power per area) on the other hand, which for xenon at atmospheric pressure was measured as 2 W/cm^2, is in the normal glow discharge mode independent of current. This is due to the fact that with increasing current the total power and the area of emission both increase linearly.[35]

The total excimer emission and the excimer intensity are strongly dependent on pressure. For xenon, the excimer power was found to peak at a pressure of 400 Torr.[33] The intensity, however, increased linearly for currents higher than 1 mA up to the highest pressure of 760 Torr, for a discharge operated in the normal glow mode.[36] This is due to the fact that the emitting plasma area decreases with pressure.

Modeling results in high pressure glow discharges showed that the application of pulses on the order of, or less than, the electron relaxation time (the time of energy equilibration between the electrons and the neutral gas) allows electron heating without considerable gas heating.[22] Experiments with xenon microhollow cathode discharges have confirmed this hypothesis.[37] By applying pulses of 20 ns duration with voltages of up to 750 V, not only the excimer emission could be increased up to 2.75 W, at an intensity of 15 W/cm^2, but also the efficiency reached values of almost 20%. Pulsed measurements in 10 ns pulsed argon discharges, on the other hand, showed that the power of a single discharge could be increased to 250 mW, but not the efficiency.

Whereas our studies have focused on xenon and argon as rare excimer gases, and gas mixtures which lead to ArF and XeCl excimer emission (Table I), Becker *et al.* have studied discharges with the same electrode geometry [Fig. 1(b)] in neon and helium and have recorded neon excimer radiation at 84 nm and even helium excimer radiation at 75 nm.[11]

IV. MICROHOLLOW CATHODE DISCHARGE ARRAYS

Most applications of microdischarges require the arrangement of these discharges in arrays. Placing them in series allows us to increase the intensity, as has been shown by placing two XeCl discharges in a tandem configuration.[38] However, generating a line of excimer sources using this method requires aligning 100 μm holes in a multiple sandwich (metal–dielectric–metal) structure, a technically difficult task using laser drilling. Using etching methods in silicon as electrode material seems to allow the manufacturing of larger systems. However, even in this case, the system was limited to three discharges in series.[39]

Placing the discharges in parallel, as required, for example, for flat panel excimer lamps, is easier. For discharge modes where the current voltage characteristic has a negative slope (hollow cathode discharge mode) or is flat (normal glow discharge mode) it is possible to generate arrays of discharges by using distributed resistive ballast. This has been demonstrated by using semi-insulating silicon as anode material.[40] If operated in discharge modes where the current voltage characteristic has a positive slope, the discharges can be arranged in parallel without individual ballast.[2] Operating in the predischarge mode requires reducing the current to small values such that space charge effects are negligible (generally <1 mA). Operation in the abnormal glow mode requires limiting the cathode surface area. For the cathode dimensions on the order of mm or less (area of the cathode outside the microhollow) used in our studies, currents on the order of 10 mA were required for abnormal glow operation.

An example of an electrode structure with limited cathode area is shown in Fig. 4 (insert). A series of 30 200 μm diam holes are placed along a line, with distances of 350 μm between hole centers. The cathode area is limited by a dielectric (alumina) to a 250 μm wide strip. The anode was placed on one side on top of the 250 μm thick dielectric. The gas was a mixture of 1.5% Xe, 0.06% HCl, 0.03% H_2, and 98.41% Ne. When a voltage of 190 V was applied, the microdischarges turned on, one after another, until the entire set of discharges was ignited (Fig. 4). When all discharges were ignited, the current–voltage characteristic turned positive since all discharges were now operating in an abnormal glow mode. The excimer intensity in the abnormal glow mode (>25 mA) increases linearly with current up to values, where

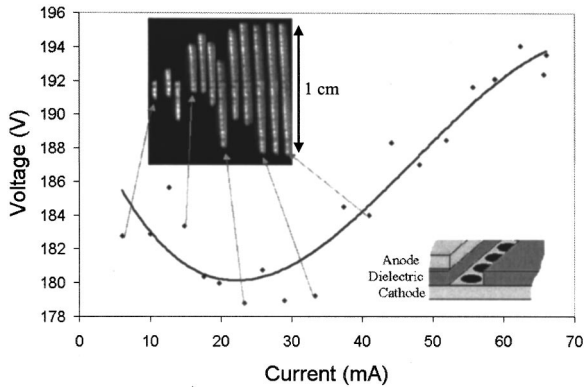

FIG. 4. Formation of a string of microhollow cathode discharges in an electrode configuration as shown in the inset. All microdischarges are ignited when they enter the abnormal glow mode, as indicated by the change in slope of the current–voltage characteristic.

the discharge transfers from glow to arc (at approximately 65 mA).

V. SUMMARY

Microhollow cathode discharges can be operated at high pressure, atmospheric pressure and greater, in a direct current mode in rare gases and in molecular gases. The sustaining voltage is only on the order of several hundred volts, and the current ranges from microamperes in the predischarge mode to amperes in the pulsed, abnormal glow mode. One of the attractive features of microhollow cathode discharges is the nonthermal electron energy distribution, which causes high ionization and excitation rates. Peak electron densities of up to $5\times10^{16}\,\mathrm{cm}^{-3}$ have been measured for pulsed atmospheric argon discharges, far exceeding those in other nonequilibrium high-pressure glow discharges. The high rate of excitation at high gas density explains the high excimer efficiency. Up to 20% internal efficiencies have been measured in pulsed xenon, but, even for dc operation, the efficiencies for all investigated gas mixtures are still in the 1%–10% range. Mixing rare gases with molecular gases allows us, in addition, to utilize energy transfer processes to convert excimer radiation into line radiation as shown for argon–oxygen mixtures. The ability to operate such discharges in modes with positive current–voltage characteristics allows us to generate simple flat panel light sources for any of the excimer gases. The thin electrode structure permits good thermal management. This would be required to fully utilize the high intensity that reaches values of 2 $\mathrm{W/cm}^2$ for dc and 15 $\mathrm{W/cm}^2$ for pulsed operation for xenon excimer emission.

A distinct feature of microhollow cathode discharges is the extreme power density in these discharge plasmas. Typical discharge voltages are approximately 200 V. Currents on the order of 1 mA are typical in such discharges when operated dc in the hollow cathode discharge mode, resulting in an electrical power for single discharges on the order of 100 mW. The volume of cathode fall and negative glow, where most of the electrical energy is dissipated, is assumed to be determined by the cathode opening. For a 100 μm thick, 100 μm diam cathode opening the volume is $0.8\times10^{-6}\,\mathrm{cm}^3$. The volume power density is consequently on the order of $10^5\,\mathrm{W/cm}^3$. This allows us to utilize such microdischarges as plasma reactors.[18] Also, with power densities that high conditions for lasing should be achievable when multiple such discharges is arranged in series.

ACKNOWLEDGMENT

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