High-efficiency, high-power, stable 172 nm xenon excimer light source

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Stable, continuous-wave light sources at 172 nm, based on the Xe_2* excimer molecule, with conversion efficiency of electrical energy to vacuum ultraviolet (VUV) light greater than 50%, are reported. In high-pressure xenon gas, ''Saint Elmo's Fire'' corona discharges serve as localized point electron sources with a metal grid at a few kilovolts providing an accelerating electric field. An extended VUV light-emitting region with high-energy conversion efficiency indicates that electron energy loss is predominantly by excitation of Xe atoms rather than by ionization. A room-temperature prototype lamp with variable VUV power to 35 mW/cm^2 has been demonstrated. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1605798]

For centuries, people have been fascinated by the glow around a charged insulated point, such as a ship's mast or a high-voltage power line under certain atmospheric conditions. This glow, a corona discharge known as Saint Elmo's fire, was described, for example, by the American novelist Herman Melville[1] in *Moby Dick*. In this work, we show how St. Elmo's fires in dense rare gases can be miniaturized, controlled, and used to generate ultraviolet light, with broad applications in science and industry. We demonstrate that point glows can be a source of electrons for excimer, rare gas excited state molecule formation. Excimers emit in the vacuum ultraviolet (VUV) region of the spectrum where the high energy per photon is especially suitable for applications in materials processing, photochemistry, analytical chemistry, sterilization, ozone production and water purification. When efficient two-photon conversion to the visible region[2] becomes practical, excimers will become useful for display and general lighting applications, too. Until now, only limited VUV light sources have been available for applications such as surface modification and cleaning of semiconductors during fabrication,[3] chemical vapor deposition (CVD) of metal films from the gas phase,[4] improvements to the structure and stoichiometry of thin films,[5] and activation of substrates to increase surface tension. Each of these applications would benefit greatly from VUV light produced more efficiently minimizing heat production and lowering costs. Other significant applications such as ozone production and water treatment require higher efficiency and versatility than are currently available.

The production of rare gas excimers requires high pressure,[6] as the radiating molecules are produced by three-body collisions, and relatively low temperature, or the excimer molecules will dissociate before relaxing and radiating. Existing lamps with maximum efficiencies near 20% are based on special, often pulsed, electrical discharges such as the dielectric barrier discharge.[7] An alternative excimer production method uses relatively low-energy electron beams and yields conversion efficiencies of pumping power to VUV-light near 40%.[8] This is close to the theoretical maximum based on energy balance including ionization and atom excitation, but has much lower overall efficiency because of the energy used to produce the electrons.

As the excitation energy of an atom is always below its ionization threshold, it may be possible to significantly increase the conversion efficiency by creating conditions where the electron can transfer enough energy to excite the atom but not enough to ionize it. This is the process employed in the mercury fluorescent lamp, where electrons are accelerated by an electric field in low-pressure mercury vapor to yield mercury ultraviolet (UV) radiation with a theoretical conversion efficiency near 60%.[9] Until now, it has not been possible to create similar conditions for excimer production because of the high pressures required, as electron production in a cold, dense gas usually requires strong electric fields, which lead to ionization as well as excitation.

Now, we have shown that a system of two electrically coupled regions in a high-pressure rare gas, a discharge electron production region, and an electron drift excimer production region, can be made to yield VUV radiating excimers with minimal ionization loss. In the first region a spatially limited corona discharge serves as a localized plasma cathode. The electrons then enter a drift region with an electric field of sufficient strength for them to gain enough kinetic energy per average collision length to excite but not to ionize atoms. The only mechanism for electron energy loss, other than excitation, is the minute impulse momentum transfer to rare gas atoms via elastic collisions.

The Saint Elmo's fire plasma cathode region, shown schematically in Fig. 1, consists of a conducting needle at a negative high voltage in a dense rare gas. A small, bright, negative corona discharge can be observed around the tip. All experiments reported here use xenon at a pressure near 1 atm emitting at 172 nm, a wavelength easily transmitted by commercial VUV grade fused silica. The electric field on the symmetry axis of the tip decreases inversely with distance[10] from the surface. As a result, the field is sharply peaked, leading to a localized electrical discharge. Outside the discharge region the electric field drops rapidly and is not strong enough to accelerate electrons generated in the point plasma region to the excitation or ionization energy of gas atoms before they collide. The electrons simply drift slowly to the nearest conductor at ground potential. A single needle dis-

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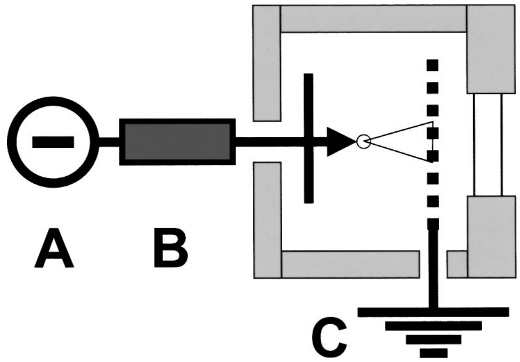

FIG. 1. Schematic drawing of a single needle corona discharge Xe_2* VUV-excimer lamp in point-to-plane geometry. (A) is the high-voltage power supply, (B) a ballast resistor ($R_B = 100\ \mathrm{M}\Omega$), and (C) the cell filled with 1.3 bar research grade xenon gas with corona needle, grid-like ground plane, and CaF_2 VUV-output window. With 4.5 kV from the power supply a current of 30 μA flowed through the lamp.

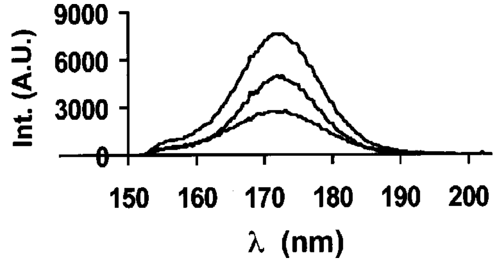

FIG. 2. Spectra of the second excimer continuum of Xe_2* at 172 nm from the decoupled source–drift system. The upper spectrum is the VUV emission of the microdischarge plasma cathode and the drift region combined. The lower spectrum is VUV emission of the microdischarge alone. The center spectrum is the difference between the two and represents the VUV emission of the drift region alone.

charge at a few thousand volts can produce up to 100 μA of electron current. With a corona needle at negative potential and the symmetry broken by a grounded planar metal grid about 1 cm from the tip, electrons generated in the point plasma region accelerate toward the grid and form a cone-shaped drift region. If the point-to-plane field is maintained at a value such that electrons acquire sufficient energy to excite but not ionize before colliding, an efficient, stable, cone-shaped excimer formation region results. The electron current generated in the plasma cathode generates Xe_2* excimer molecules, and consequently VUV light, in the drift region. Furthermore, the high excitation efficiency in the drift region more than compensates the inefficiency of electron generation in the discharge region.

If an energy ε_e greater than the ionization energy $\varepsilon^{\mathrm{ion}}(\mathrm{Xe})$ is required to create one electron in the point corona discharge and an electron makes N inelastic collisions with sufficient energy ε^* to produce N excited xenon atoms in the drift region, each yielding a VUV photon with average xenon excimer photon energy 7.22 eV, then the total excimer light production efficiency η is the ratio of VUV photon energy emitted to the energy transferred to the electron:

$$\eta = \frac{N 7.22\ \mathrm{eV}}{N\varepsilon^* + \varepsilon_e} = \frac{7.22\ \mathrm{eV}}{\varepsilon^*}\,\frac{1}{1 + \dfrac{\varepsilon_e}{N \cdot \varepsilon^*}}.$$

For Xe, $\varepsilon^* = 8.32$ eV,[11] and for $N\varepsilon^* \gg \varepsilon_e$, the efficiency of the entire system may reach 87%.

To investigate the drift region more closely, we replaced the corona needle with a cw microdischarge between two closely spaced needle tip electrodes. The potential between the microdischarge plasma cathode and a metal mesh ground plane was chosen such that only about 1% of the microdischarge current would enter the drift region. Spectra are shown in Fig. 2. The upper spectrum shows the second continuum of Xe_2* produced by the microdischarge and the drift region together. The lower spectrum is that of the microdischarge alone with an input electrical power of 125 mW. The center spectrum is the difference and represents the VUV emission of the drift region, where the product of extracted current and accelerating voltage was only 12 mW. The wavelength integrated intensity from the drift region constitutes 62% of the total VUV emission of the combined system. As a result, the electrical pumping power to VUV light conversion efficiency of the drift region can be estimated to be about 17 times higher than the efficiency of the micro glow discharge alone. The VUV power emitted was measured using a calibrated photodetector.[12] An efficiency of up to 54% was measured for the drift region, while the microdischarge only yielded 3%, a value comparable to efficiencies reported for microhollow cathode discharges.[13]

The full width at half maximum in nm of the second excimer continuum can be used to estimate the temperature T of the light-emitting region.[14] The temperature estimate for the microdischarge region is 770 ± 40 K and for the drift region 360 ± 30 K, where a cold gas environment favors excimer formation leading to higher efficiency.

In order to operate this two-region excimer light source in a stable and efficient manner, the electric field in the drift excitation region and the system pressure had to be carefully optimized. At a given pressure exceeding a maximum field led to a breakdown. At low fields there is insufficient energy for excitation between collisions. A detailed model of the drift region efficiency based on the derivation of the spatially varying electron energy distribution function, yields good agreement with the experimental results.[15]

Two prototype devices using corona needle tip plasma cathodes were constructed. One used a single tungsten needle corona tip in point-to-plane geometry as in Fig. 1. Measurement of the VUV-light output taking the solid angle subtended by the detector into account yielded conversion efficiencies of up to 55%, for the system of corona discharge plus drift region combined.

The second prototype used 21 point-to-plane corona discharges in a large-area configuration. The parameters for the lamp (Fig. 3) were: Xe pressure greater than 2 bar; 21 needles, 6 mm apart, with approximate radii of curvature 5 μm; 2.4 cm to a grounded wire mesh with more than 95% open area; and 4 mm to a 5-cm-diam UV-enhanced fluorine-doped modified silica[16] output window. Each needle is isolated by a thin glass tube from a flat, polished aluminum disk cathode held at negative high voltage. The polished surface serves as a reflector for VUV. By varying the voltage applied to the lamp, the VUV output power could be varied from 0 to more than 35 $\mathrm{mW/cm^2}$ at the center of the output window, with a conversion efficiency of 20%. The spectral output of the lamp was similar to the center spectrum of Fig. 2.

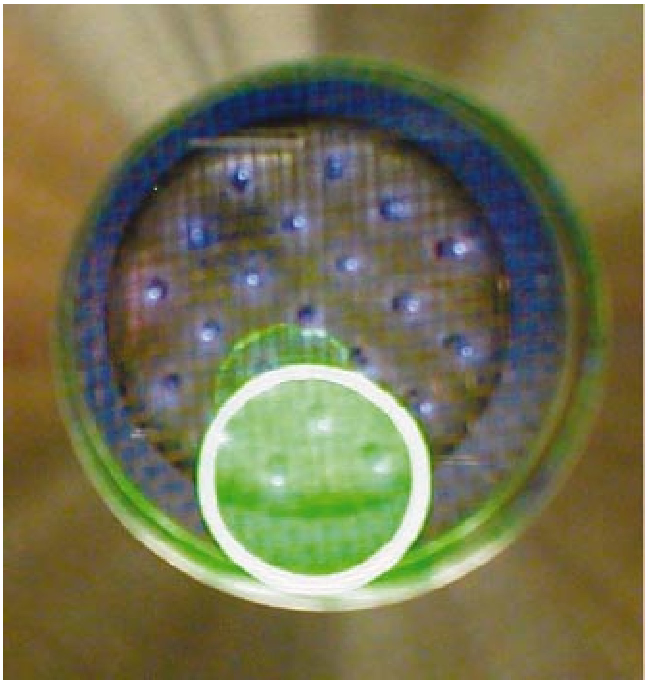

FIG. 3. (Color) Photograph of highly efficient cw prototype large area VUV Xe_2* excimer lamp consisting of 21 corona needles in parallel showing St. Elmos's fires and green fluorescence of UV-sensitive glass.

Similar, highly efficient (>50%) larger area cw 172 nm VUV Xe_2* excimer lamps operating at room temperature with output power exceeding 200 mW/cm^2 should be possible to construct using larger lamp diameters. Due to the high conversion efficiency of electrical energy into VUV excimer radiation, the gas and the lamp itself require no cooling and remain basically at room temperature. The 172 nm lamp should find immediate application in UV cleaning and as an efficient device for ozone production. At 172 nm there is essentially 200% photon efficiency for ozone production in oxygen or air, as nitrogen is unaffected by 172 nm radiation. Similar lamps at other wavelengths might also be constructed with other excimer systems.

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