

Neutral bremsstrahlung measurement in an atmospheric-pressure radio frequency discharge

Jaeyoung Park,^{a)} Ivars Henins, Hans W. Herrmann, and Gary S. Selwyn
Plasma Physics Group, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

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Neutral bremsstrahlung emission spectrum is measured in an atmospheric-pressure radio frequency (rf) capacitive discharge for a gas mixture of helium (99.5%) and oxygen (0.5%) using a high resolution triple monochromator between 450 and 1000 nm. Good agreement is obtained for spectral variation and absolute intensity between the observed neutral bremsstrahlung and theoretical emissivity calculated using electron–neutral momentum cross sections. Based on a theoretical fitting, the discharge is characterized by a time averaged electron density of $2.9 \times 10^{11} \text{ cm}^{-3}$ and an electron temperature of 1.9 eV for an input power density of 28 W/cm^3 .

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Continuum emission and its inverse process of continuum absorption has been studied extensively in ionized gas media such as stellar atmospheres, arcs, shock tubes, and thermonuclear fusion reactors. In almost all of these media, continuum emission originates from free electrons undergoing sudden changes in velocity under the influence of Coulomb fields of other charged particles and neutrals, present in the media. Thus, the continuum emission generally contains valuable information on the statistical state of the free electrons, such as the electron density and temperature which is often difficult to measure by other means. Many studies on the various mechanisms for continuum emission (and absorption) have been conducted in plasma physics and astrophysics, establishing the continuum emission measurement as a standard means to understand the ionized gas state.^{1,2} Of the different mechanisms for continuum emission, however, the theoretical calculation for neutral bremsstrahlung ($e + A \leftrightarrow e + A + h\nu$, where A is the atom) has been considered more difficult compared to others due to the multi-body nature of the underlying interaction. Moreover, few experimental measurements, with exceptions being the works by Taylor and Caledonia³ and by Kung and Chang,⁴ have been made exclusively for neutral bremsstrahlung emission spectrum over a wide range of wavelengths. Thus, the accuracy of various theoretical calculations, each based on different approximations, has not been examined thoroughly.

In this paper, we report an experimental measurement of plasma emission in an atmospheric-pressure rf capacitive discharge, where neutral bremsstrahlung is the dominant source of the continuum emission. The emission spectrum is measured using a triple monochromator, which separates the continuum emission from molecular bands and atomic lines and excludes stray light in the spectrometer. The obtained spectrum shows good agreement with the theoretical formula by Dalgarno and Lane,⁵ which uses experimentally measured momentum cross sections for calculating neutral bremsstrahlung emissivity.

A schematic of the experimental setup is shown in Fig. 1. The discharge is produced between the two parallel aluminum plates using rf electric fields at 13.56 MHz. Each electrode is 1.5 by 10 cm, with a gap spacing of 0.24 cm. This source is capable of producing a stable, (oscillating) steady-state discharge at atmospheric pressure. Nominal discharge parameters are electron densities of 10^{10} – 10^{11} cm^{-3} , electron temperatures of 2–4 eV and gas temperatures of 50–300 °C. In this parameter regime, neutral bremsstrahlung emission dominates electron–ion bremsstrahlung emission as the neutral gas density exceeds the density of charged particles by several orders of magnitude. Detailed descriptions of the discharge source can be found in previous studies.^{6,7}

As shown in Fig. 1, the collection optics for the continuum emission measurement consists of a collimating slit assembly, a quartz fiber bundle and a triple monochromator with a liquid nitrogen cooled charge-coupled device (CCD) array detector. The slit assembly of two 8 mm by 83μ slits, separated by 2.5 cm, was used to accurately limit the light collection volume of the discharge and to prevent scattered light from the electrode surfaces from entering the fiber bundle. To suppress stray light in the spectrometer and to provide adequate spectral resolution (0.3 nm), a triple monochromator (SPEX 1877 Triplemate) was used with a 1200 groove/mm grating in the final stage. The continuum emission intensity was examined for stray light at several wavelengths using varying bandwidths in the filter stage, from 2 to 30 nm. No change in the continuum intensity was observed.

The absolute calibration of the observed plasma emission intensity was conducted by replacing the discharge source with a pinhole (400 μ diam) image of a standard tungsten ribbon filament lamp. The day-to-day variation in the emission intensity is about 5% at 700 nm, while the uncertainty in absolute calibration is less than 10% for spectral intensity variation and about 15% for absolute intensity. Note that the emission spectrum was measured from 450 to 1000 nm. This is because the efficiency of this triple monochromator drops rapidly in the near ultraviolet (UV) region because all three gratings are blazed for about 650 nm. In

^{a)} Author to whom correspondence should be addressed. Electronic mail: jypark@lanl.gov

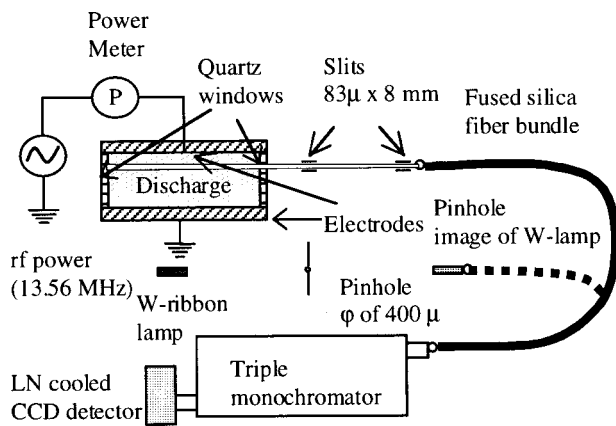


FIG. 1. A schematic of the experimental setup showing the discharge source and the collection optics.

addition, the sensitivity of the CCD decreases rapidly above 1000 nm.

In Fig 2, a raw spectrum from the mid-section of the discharge is shown before calibration. The averaging along the line of sight is not an issue due to the limited light col-

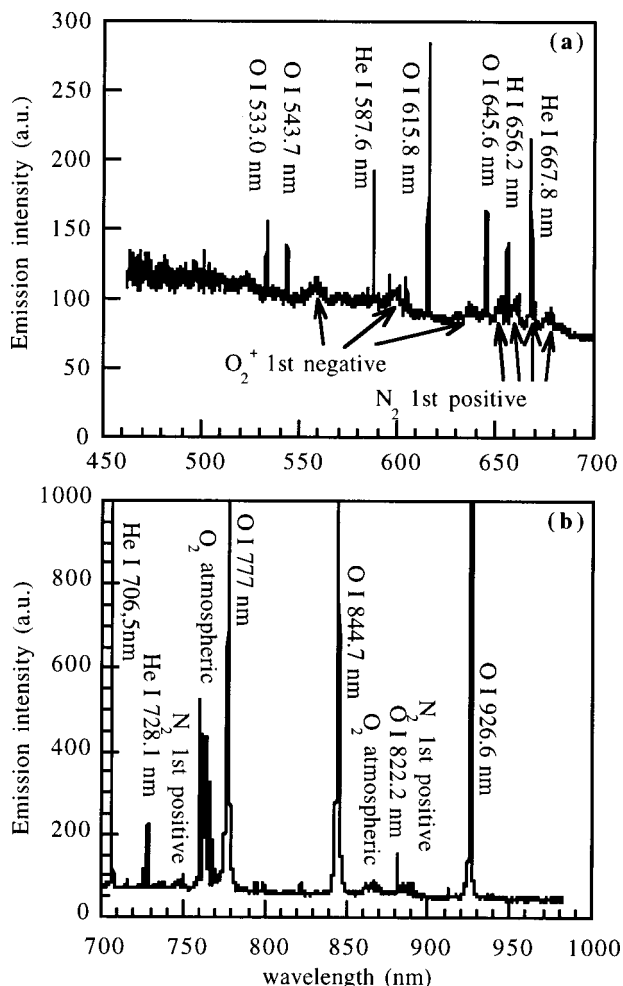


FIG. 2. A sample raw emission spectrum taken from the mid-section of the discharge. The discharge condition is: input power of 100 W (equal to 28 W/cm³) and gas flow rate of 25 slpm using a gas mixture of helium (99.5%) and oxygen (0.5%).

lection volume (160 μ in vertical dimension) as the emission intensity changes less than 20% within 0.05 cm from the center of the gap. The discharge condition is: Input power of 100 W (equal to 28 W/cm³) and a gas flow rate of 25 slpm using a gas mixture of helium (99.5%) and oxygen (0.5%). The spectrum exhibits strong continuum emission, clearly distinguished from atomic lines and molecular band structures. Prominent atomic lines are from oxygen neutral atoms (616, 777, 845, and 927 nm) and helium neutrals (588, 668, 706, and 728 nm). No line emission from ions are observed, consistent with a very low ionization fraction. In addition, two oxygen molecular band systems are observed, i.e., O₂ atmospheric system (760 and 860 nm) and O₂⁺ first negative system (642, 603, and 563 nm). The relatively strong nitrogen first positive system is also observed in several wavelength regions.

The presence of nitrogen band emission is probably due to the discharge source being only airtight, thus allowing a small leakage of air into the discharge region. In fact, the main reason for the addition of oxygen is to reduce the intensity of the nitrogen bands, which otherwise clutter the spectrum. With 0.5% addition of oxygen, the strongest nitrogen band intensity is less than 25% of the continuum emission intensity without apparent overlap of different band systems. At this small fraction, the contribution of oxygen molecules and atoms to neutral bremsstrahlung emission can be neglected when compared to that of helium atoms.⁵

Furthermore, the emission spectrum was monitored at three different wavelength regions (at 500, 700, and 900 nm) as a function of nitrogen concentration by adding small quantities of nitrogen to the gas mixture of helium and oxygen. This procedure was performed to eliminate the possibility of NO emission, which is known to be continuous in the visible range, and to ensure that the measured continuum is free from any band emission.⁸ At 0.12% nitrogen fraction, the continuum emission intensity changes less than 5% while the intensity of N₂ first positive system increases by a factor of 10 or more, compared to the results with no addition of nitrogen. This result eliminates the possibility of NO emission and provides an estimate for the experimental error (5%) in neutral bremsstrahlung intensity resulting from molecular bands not sufficiently resolved by the spectrometer.

To analyze the observed neutral bremsstrahlung emission, the analytical formula for neutral bremsstrahlung emission cross section by Dalgarno and Lane, Eq. (1), is used based on the literature survey by Johnston,⁹

$$\frac{d\sigma_{\nu}(E)}{d\nu} = \frac{8r_e}{3c} \frac{E}{h\nu} \left(1 - \frac{h\nu}{E}\right)^{1/2} \left[q_0(E-h\nu) + \left(1 - \frac{h\nu}{E}\right) q_0(E) \right] \text{ in cm}^2 \text{ s}, \quad (1)$$

where r_e is the classical electron radius, c is the speed of light, E is the initial electron energy, $h\nu$ is the photon energy, and q_0 is the electron momentum cross section as a function of electron energy. This formula expresses the neutral bremsstrahlung cross section in terms of electron-neutral elastic scattering cross section in the limit of low-energy photons using the phase-shift approximation.¹⁰⁻¹⁴ As will be

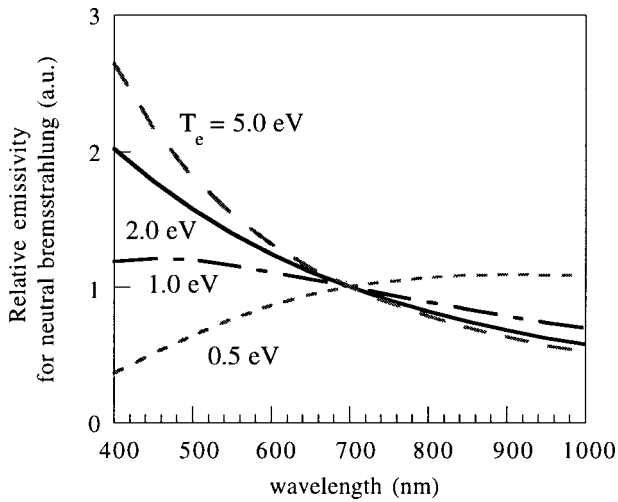


FIG. 3. Relative neutral bremsstrahlung spectral emissivity as a function of electron temperature. See the text for the absolute values.

shown later, the Maxwellian electron temperature in the discharge is about 2 eV, thus justifying this approximation. In the limit of high electron energy ($E \gg h\nu$), this formula is reduced to the semiclassical result by Zel'dovich and Raizer.¹⁵ The use of this formula is attractive for our study, as the accuracy of the momentum cross section of helium is better than 10%.

The spectral emission coefficient for neutral bremsstrahlung is calculated by integrating Eq. (1) over the electron energy distribution,

$$Q_\lambda d\lambda = N_{\text{gas}} N_e h \nu \left(\int_{h\nu}^{\infty} v \frac{d\sigma_\nu(E)}{d\nu} \frac{d\nu}{d\lambda} f(E) dE \right) d\lambda$$

in $\text{W/cm}^3 \text{ nm}$, (2)

where Q_λ is the energy radiated from neutral bremsstrahlung per unit time and per unit volume in the wavelength interval of $d\lambda$, N_{gas} , N_e is the density of neutral atoms and electrons, v is the initial velocity of the electron and $f(E)$ is the electron energy distribution in the gas medium. Equation (2) is then numerically integrated using the momentum cross section in the literature¹⁶ and a Maxwellian distribution with an electron temperature of T_e . Figure 3 shows the result of this calculation as a function of Maxwellian electron temperature. To emphasize the spectral variation of the emissivity, we normalized the spectral emissivity to unity at 700 nm. The absolute values of the emissivity per unit electron and gas density are 0.075, 1.51, 13.4, and 119.1 in the unit of $10^{-38} \text{ W/cm}^3 \text{ nm}$ at 700 nm for T_e of 0.5, 1.0, 2.0, and 5.0 eV, respectively. Note that $Q_\lambda (=Q_\nu/c/\lambda^2)$ is calculated rather than Q_ν for later comparison with the experimentally measured emission intensity. The use of a Maxwellian distribution should be reasonable since the probability of emitting a photon does not vary rapidly with the initial electron energy as long as the electron energy is above the photon energy. Thus, for the wavelength region in this paper (1000–450 nm, or 1.2–2.8 eV), the major contribution in Eq. (2) comes from the relatively low energy part of the distribution where it can be reasonably approximated by a Maxwellian distribution.¹⁷

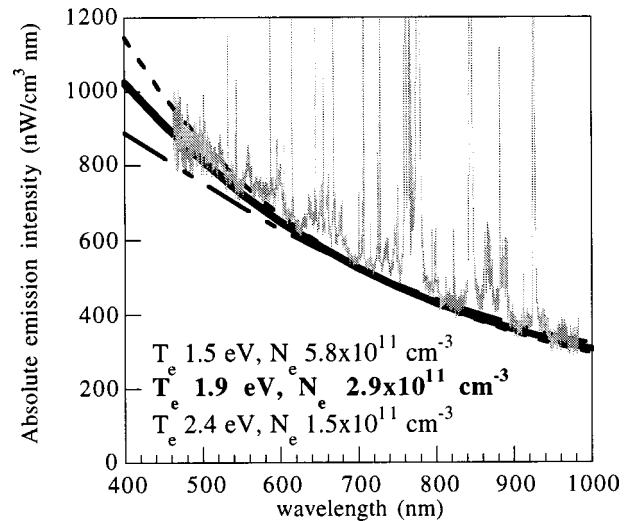


FIG. 4. Comparison between the observed absolute plasma emission intensity, calibrated from Fig. 2, and three theoretically calculated neutral bremsstrahlung emissivity for electron temperatures of 1.5, 1.9, and 2.4 eV and electron densities of $5.8, 2.9, \text{ and } 1.5 \times 10^{11} \text{ cm}^{-3}$, respectively.

In Fig. 4, the observed emission spectrum after absolute calibration is compared with the calculated neutral bremsstrahlung emissivity. The fitting is performed in two steps. First, the relative spectral variation of the observed continuum emission spectrum and the calculated bremsstrahlung emissivity are compared to yield a Maxwellian electron temperature. This is followed by adjusting the absolute intensity of the calculated emissivity to match the observed continuum emission intensity to yield an electron density. It is noted that the neutral gas density of $1.6 \times 10^{19} \text{ cm}^{-3}$ for a gas pressure of 600 Torr (atmospheric pressure in Los Alamos, NM) is calculated from the ideal gas law using the measured rotational temperature (100°C) of gas, derived from O_2 atmospheric band emission intensity at 760 nm.⁷ The fit results in a Maxwellian electron temperature of $1.5 \text{ eV} < T_e < 1.9 \text{ eV} < 2.4 \text{ eV}$ and an electron density of $1.5 \times 10^{11} \text{ cm}^{-3} < N_e < 2.9 \times 10^{11} \text{ cm}^{-3} < 5.8 \times 10^{11} \text{ cm}^{-3}$. It should be noted that the results are time averaged values over an rf period.

This electron temperature of 1.9 eV agrees with our previous estimate of electron temperature of 2.0 eV obtained from average electric field strengths based on experimental measurements for similar experimental parameters.⁷ In addition, the estimated electron temperature and density are consistent with a simple power balance between the electron heating by the rf fields and the electron energy loss by collisions with neutrals in Eq. (3),

$$P_{\text{in}} \sim P_{\text{loss}} \approx N_e \left(\frac{3}{2} k T_e - \frac{3}{2} k T_n \right) 2 \frac{m_e}{m_{\text{He}}} \nu_{\text{en}} \text{ in } \text{W/cm}^3, \quad (3)$$

where P_{in} is the input power density, m_e/m_{He} is the mass ratio between the electron and helium atom, and ν_{en} is the collision frequency of elastic scattering for electrons with helium atoms. For the electron temperature of 1.9 eV and the density of $2.9 \times 10^{11} \text{ cm}^{-3}$, the left-hand-side (lhs) of Eq. (3) becomes 20 W/cm^3 for ν_{en} of $5.7 \times 10^{11} \text{ Hz}$, consistent with the input power density of 28 W/cm^3 .

Based on the agreement discussed above, it is concluded that the neutral bremsstrahlung emissivity can be calculated with reasonable accuracy in the case of helium using experimentally measured momentum cross sections. However, further experiments are needed to verify the same theoretical approach to other gases. In particular, the early works by Taylor and Caledonia, and Kung and Chang, reported that a large discrepancy (a factor of 10) may exist between the observed emissivity and the theoretical calculation in the case of argon and xenon due to the Ramsauer effect.¹⁸ Finally, this study shows that the neutral bremsstrahlung may be used to measure both electron density and temperature in certain types of ionized gas media, especially those in which no other experimental means are currently available to directly measure these quantities. Examples of this include high-pressure plasma sources, such as the high-pressure rf source for gas laser generation and plasma display panel discharges.

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