

Spectroscopic temperature measurements of air breakdown plasma using a 110 GHz megawatt gyrotron beam

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Temperature measurements are presented of a non-equilibrium air breakdown plasma using optical emission spectroscopy. A plasma is created with a focused 110 GHz 3 μ s pulse gyrotron beam in air that produces power fluxes exceeding 1 MW/cm². Rotational and vibrational temperatures are spectroscopically measured over a pressure range of 1–100 Torr as the gyrotron power is varied above threshold. The temperature dependence on microwave field as well as pressure is examined. Rotational temperature measurements of the plasma reveal gas temperatures in the range of 300–500 K and vibrational temperatures in the range of 4200–6200 K. The vibrational and rotational temperatures increase slowly with increasing applied microwave field over the range of microwave fields investigated. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4773037>]

I. INTRODUCTION

While DC to low frequency microwave air breakdown has been extensively studied,^{1,2} relatively little attention has been paid to high frequency microwave discharges. This omission is mainly due to a historical lack of high power source technology at millimeter and sub-millimeter wavelengths. The development of high power, high frequency sources such as gyrotrons has made it possible to create large volume electrodeless plasmas in atmospheric pressure air at microwave frequencies greater than 100 GHz. In multiple experiments, these pulsed plasmas have been shown to possess unique geometries and complex dynamics not seen at lower frequency.^{3,4} In particular, the observation of plasma filaments aligned with the electric field polarization that propagate back toward the microwave source at $\lambda/4$ intervals has been documented. Much is still unknown about the dynamics and plasma parameters of these unique features, and an interest to better explain high frequency microwave breakdown has motivated further investigation of plasmas created by a high power gyrotron.

In order to better understand high frequency microwave breakdown, it is necessary to investigate the parameters characterizing the plasma as has been done for many low frequency and DC breakdown experiments.^{5–8} Plasma temperatures and plasma constituent concentrations are important to the overall spatial evolution of the plasma. Detailed measurements of these parameters enable a better understanding of the dynamics of plasma streamer evolution and give insight to the chemical processes occurring in the plasma. In particular, it is of interest to study the plasma parameters as they vary with applied field and pressure, as these two variables are important in any future application of high frequency microwave breakdown and also have been shown to have a significant effect on breakdown at low frequency both in theory and experiment.⁹

Optical emission spectroscopy (OES) is widely used as a plasma diagnostic and permits the investigation of plasma parameters without the use of invasive probes that can perturb the behavior of the plasma. OES has been used to measure excitation, rotational, vibrational, and electron temperatures, as well as electron density and the concentration of plasma species in low temperature, non-equilibrium air discharges.^{10–12} There has been a limited amount of research using spectroscopic techniques to measure the plasma parameters of air and nitrogen breakdown using gyrotrons.^{13,14} In particular, OES has been used to measure electron density and temperature when equilibrium models were assumed for discharges using a focused beam from a gyrotron in nitrogen.¹⁵ However, to our knowledge, no measurements of plasma parameters using OES in non-equilibrium air plasmas using a gyrotron have been previously reported. OES measurement techniques do not perturb external electromagnetic fields and so can be used in situations where plasma dynamics are fast or electric fields are strong and are crucial to the spatial evolution of the plasma, as is the case in streamer formation in high frequency microwave breakdown.

A. Low temperature plasma optical emission spectroscopy

Plasma temperatures are important in air discharges because they define the thermal and chemical state of the plasma. In general, several different temperatures are used to characterize plasmas that are not in thermal equilibrium. For a plasma formed in a monatomic gas only the translational and electronic energy of the atoms are of importance in determining the emission spectra. Electronic transitions give rise to the spectral lines observed and line broadening is due to the translational energy of the atoms. The population distribution of electronic and translational energy states defines the excitational and gas temperatures, respectively. However, for a plasma created in a molecular gas the total energy of the plasma must also include the relative motions of the

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atoms comprising the molecules. For a gas such as air where the majority of the constituents are diatoms, the total energy of any given molecule also includes the energy corresponding to the vibrational and rotational energy of the two atoms with respect to one another. In any one electronic transition, there may also be a transition between rotational or vibrational states as long as selection rules are followed. These added degrees of freedom in general have a significant effect on the observed spectra. For each of the different degrees of freedom a temperature is defined with the associated population distribution of states in that degree of freedom: T_{rot} for rotational, T_{vib} for vibrational, T_{exc} for electronic, and T_{gas} for the translational. In addition to these temperatures is the electron temperature, T_e , which describes the continuous distribution of free electron energies.

B. Air breakdown

In gas breakdown, the critical field required to cause an electrical discharge, E_{crit} , is typically defined as the minimum field necessary to cause an electron avalanche process that raises the free electron density several orders of magnitude over the background in a short time τ ($n/n_0 \approx 10^8 - 10^{12}$). Experiments have been performed to determine E_{crit} as a function of pressure at many different frequencies in air and other gases.¹ For microwave experiments, this field has been shown to follow a Paschen-like curve similar to that observed in DC experiments, and the general form of this curve can be explained by the balance between pressure dependent electron loss processes and ionization, which is also pressure dependent. The critical field for air, expressed analytically by Taylor *et al.*¹⁶ by using an empirical fit to ionization rates, is given by the expression

$$E_{crit} = 375 \times p \sqrt{1 + \frac{\omega^2}{\nu_c^2} \left(\frac{D}{p\Lambda^2} + 6.4 \times 10^4 \right)^{3/16}}, \quad (1)$$

where the above field is in V/m, D is the electron diffusion coefficient in m^2/s , p is the pressure in Torr, ν_c is the electron-neutral collision frequency (usually taken as $\nu_c = 5.3 \times 10^9 p$ in air), and Λ is the characteristic diffusion length in meters. There is therefore an interest to understand how the plasma parameters behave as the electric field is increased above this threshold, and this is the focus of this paper.

II. EXPERIMENT

The megawatt gyrotron experimental setup with spectrometer is shown in Fig. 1. Air breakdown is initiated using a quasioptical beam from a pulsed 110 GHz MW gyrotron that is focused in a pressure chamber of flowing room air. The gyrotron pulse length is $3 \mu\text{s}$. A polypropylene beam splitter and an RF diode were used to monitor and record the gyrotron output power. The beam is focused with a 14 cm focal length HDPE lens, producing a peak irradiance of $1.5 \text{ MW}/\text{cm}^2$ in the focal spot and a diffraction limited spot size radius of $w_0 \approx 4 \text{ mm}$. The chamber pressure as well as the output power of the gyrotron beam are varied, while the

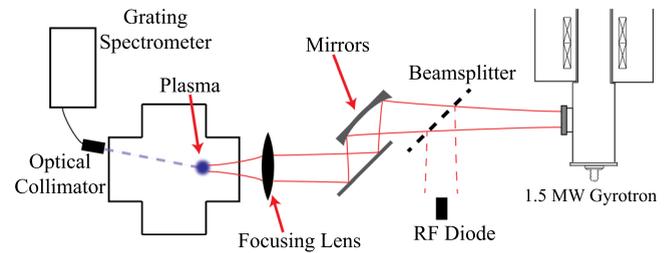


FIG. 1. Simplified schematic of experimental setup showing 110 GHz gyrotron beam and focusing optics, as well as the optical collimator alignment.

optical emission from the plasma is recorded. The breakdown plasma is several centimeters in size and emits in the visible and UV spectrum. At atmospheric pressure, the emission is not uniform, but comes from a filamentary array of streamers that propagate along the electric field polarization and back towards the gyrotron. However, it has been observed that for pressures less than 100 Torr the plasma emission is fairly uniform¹⁷ and the resulting spatial gradients associated with plasma density and temperature are likely much smaller than at atmospheric pressure if they follow the emission intensity.

A fused silica collimating lens was used to focus the emitted light from the plasma onto an optical fiber with transmission between 300 and 1100 nm. The collimator is aligned to collect light along the full length of the breakdown. Because the population of upper energy states falls off very rapidly due to their Boltzmann distribution, we expect the optical emission spectrum to be dominated by the hot, central core of the plasma and not the cooler, thin edge of the plasma even though the emission intensity is integrated along the full length of the discharge. The emission spectrum was monitored with two different grating spectrometers. A 0.64 m spectrometer with a resolution of $\sim 0.03 \text{ nm}$ was used in conjunction with a fast gating Andor ICCD camera as a detector to monitor the rovibrational lines of nitrogen. The emission spectrum was integrated over the full lifetime of the breakdown, which is about $2 \mu\text{s}$. An Ocean Optics QE 65000 spectrometer with a resolution of $\sim 1 \text{ nm}$ and a detector wavelength range of 200–1100 nm was used to collect broadband emission spectra over the UV-Vis wavelength range and identify emitting species. In Fig. 2, a typical emission spectrum obtained at an incident intensity of $1.5 \text{ MW}/\text{cm}^2$ at 1 Torr is shown. Strong emission from the nitrogen second positive system is observed from 300 to 500 nm, while the nitrogen first positive and other atomic emitters are dominant for longer wavelengths.

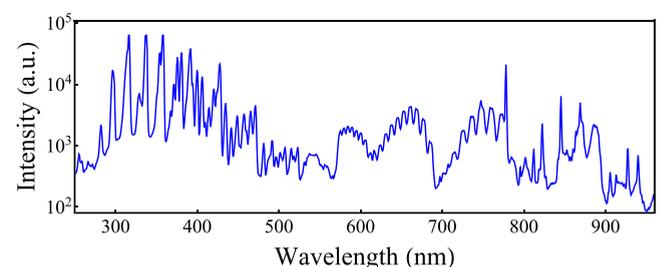


FIG. 2. Measured emission spectrum intensity on the log scale taken at a gyrotron intensity of $1.5 \text{ MW}/\text{cm}^2$ at 1 Torr using an Ocean Optics QE 65000 spectrometer.

The peak electric field and power density seen by the plasma are calculated by using Gaussian optics in the focal spot region of the 14 cm lens. The gyrotron beam has been previously measured to have a 96% Gaussian content.¹⁸ The critical field for breakdown for our experiment has been measured and shown to be in good agreement with Eq. (1), taking $\Lambda \approx w_0/\pi$ as in Ref. 17. In Fig. 3, the ratio of electric field to the E_{crit} of Eq. (1) is shown for the available gyrotron power of 1.5 MW/cm^2 over the pressure range of interest to this paper. Using this curve and knowledge of the incident gyrotron power measured with an RF diode, we record how the emission spectra and plasma parameters change as the field is increased above threshold at various pressures.

III. RESULTS

The 0.64 m spectrometer was used to record the emission spectra of molecular nitrogen. The code SPECAIR¹⁹ was used to simulate the emission spectra and compare it with the measured spectra to obtain the vibrational and rotational temperatures. SPECAIR is a line-by-line radiation code that models molecular transitions of NO, N₂, N₂⁺, O₂, CN, OH, NH, C₂, and CO, in addition to atomic lines of N, O, and C. SPECAIR does not assume LTE, but calculates the emission spectra by using user specified excitation, vibrational, rotational, and gas temperatures which are all free to vary independently. The code calculates transition rates from tabulated data and then uses these transition rates to compute the line-by-line emission intensity for each transition.²⁰ Agreement between the code's predicted spectra and spectra from low temperature air plasmas has already been reported, and the code has been used for non-equilibrium plasmas from nearly room temperature to several eV.^{11,21} SPECAIR models the finite resolution of the spectrometer by using a user defined slit function to calculate the resulting line shapes of the transitions. In addition, SPECAIR can model the optical thickness of the plasma. However, the SPECAIR best-fit to our experimental spectra showed that the plasma was optically thin.

The two temperatures T_{rot} and T_{vib} are measured by comparing SPECAIR predicted emission spectra with measured emission spectra from the $C^3\Pi_u - B^3\Pi_u$ second positive system of molecular nitrogen. This system is chosen to measure the temperature because it is very intense compared

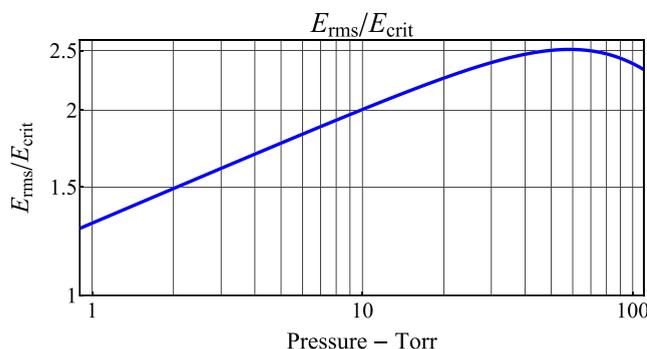


FIG. 3. E_{rms} normalized to the calculated critical rms field E_{crit} required for breakdown based on the theory by Taylor *et al.*¹⁶ given in Eq. (1) for a gyrotron intensity of 1.5 MW/cm^2 .

to emission observed from other chemical species in the air discharge. In addition, this system is commonly used to measure molecular temperatures in air breakdown and frequently observed in atmospheric plasmas.²² SPECAIR calculates emission spectra using values for T_{exc} , T_{vib} , T_{rot} , and T_{gas} . For atmospheric pressure plasmas the rotational relaxation time is very fast and the approximation that $T_{rot} \approx T_{gas}$ is taken.^{19,23} In addition, because of the fact that all measured spectra are from the same electronic transition, the relative intensities of rovibrational lines are independent of T_{exc} . Thus, the only free parameters that determine the emission spectrum generated by SPECAIR are T_{rot} and T_{vib} . The relative intensities of vibrational bandheads are used to determine the vibrational temperature, and the relative intensities of rotational lines for a given vibrational transition are used to determine the rotational temperature.

For the T_{vib} measurement, the relative intensity of the (0,1) vibrational bandhead at 357.7 nm and the (1,2) vibrational bandhead at 353.7 nm of the $C^3\Pi_u - B^3\Pi_u$ second positive system of nitrogen is compared to the SPECAIR code for a best-fit match. A typical best-fit match of the measured emission spectra and the SPECAIR code is shown in Fig. 4(a). We observe good agreement between the code and measured intensity over several orders of magnitude of intensity. The SPECAIR code is matched to emission spectra for various pressures and gyrotron powers and the best-fit value of T_{vib} for each of these cases is recorded. Then T_{vib} is plotted as a function of applied microwave electric field for five of the different pressures examined, and this is shown in Fig. 5.

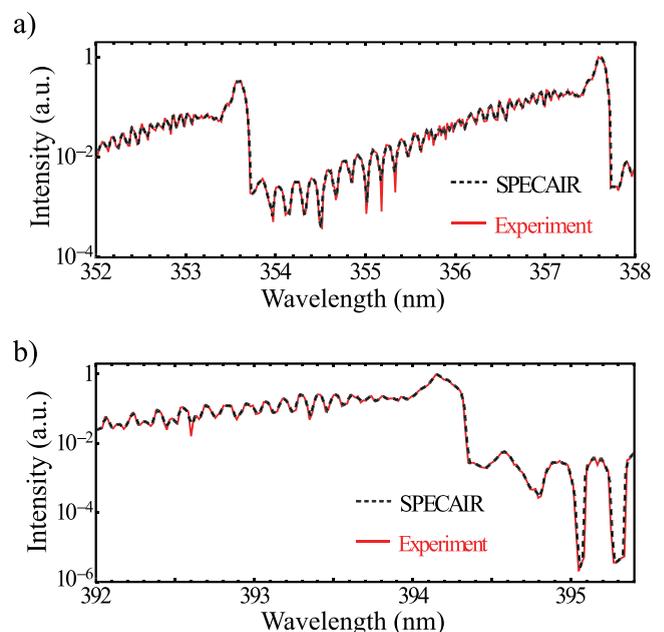


FIG. 4. (a) Measured emission spectrum of the (0,1) vibrational bandhead at 357.7 nm and the (1,2) vibrational bandhead at 353.7 nm of the $C^3\Pi_u - B^3\Pi_u$ second positive system of nitrogen compared with a SPECAIR generated best fit emission spectrum. The match gives $T_{vib} = 5000 \text{ K}$ and $T_{rot} = 450 \text{ K}$. (b) Measured emission spectrum of the (2,5) vibrational bandhead the $C^3\Pi_u - B^3\Pi_u$ second positive system of nitrogen at 394.3 compared with a SPECAIR generated best fit emission spectrum. The match gives $T_{rot} = 490 \text{ K}$. Both spectra were taken at a gyrotron intensity of 1.5 MW/cm^2 at 30 Torr.

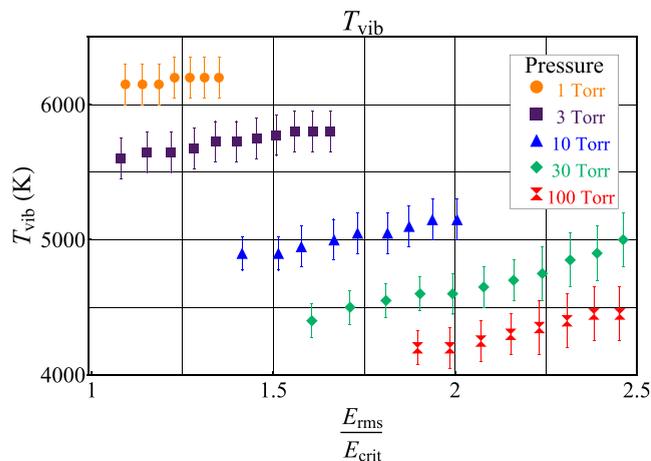


FIG. 5. Measured T_{vib} as a function of applied microwave electric field for five different pressures by comparing nitrogen vibrational bandheads at 353.7 nm and 357.7 nm with SPECAIR code.

The rotational temperature is found by matching the SPECAIR code to the relative intensities of rovibrational lines of the (0,1) vibrational bandhead at 357.7 nm. This process is also repeated for rovibrational lines of the (2,5) vibrational bandhead at 394.3 nm. These two transitions were chosen for their isolation from other transitions. Typical measured spectra vs. SPECAIR best-fit matches are shown in Figs. 4(a) and 4(b). The measured rotational temperature is shown for five different pressures as a function of applied microwave electric field for the 357.7 nm and 391.4 nm bandheads in Fig. 6 and 7, respectively. Error in the measurement of each T_{vib} and T_{rot} value is determined by the range of allowable SPECAIR temperature values that provide a match to each experimental spectrum. Error bars for the measured T_{vib} values are shown in Fig. 5, while error bars of $\pm 15\%$ in the measured values of T_{rot} have been omitted for clarity in Figs. 6 and 7.

Rotational temperature measurements made with the rovibrational lines of the (0,1) and (2,5) vibrational transitions of the nitrogen second positive system show good agreement with one another. The molecular temperature

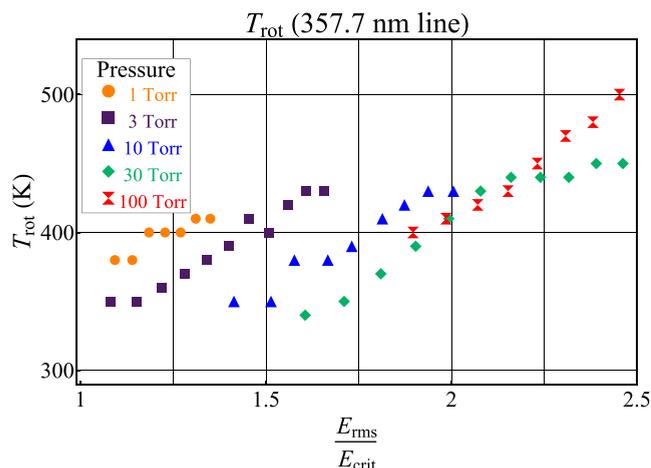


FIG. 6. Measured T_{rot} as a function of applied microwave electric field for five different pressures by comparing relative intensities of rotational lines of the (0,1) vibrational bandhead at 357.7 nm. Error bars of $\pm 15\%$ for each data point are omitted for clarity.

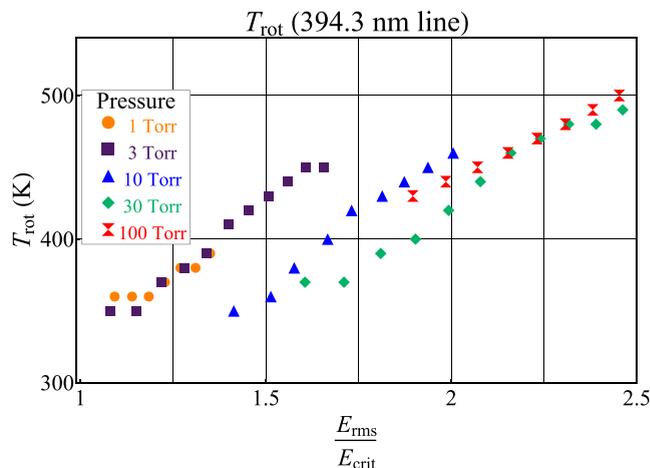


FIG. 7. Measured T_{rot} as a function of applied microwave electric field for five different pressures by comparing relative intensities of rotational lines of the (2,5) vibrational bandhead at 394.3 nm. Error bars of $\pm 15\%$ for each data point are omitted for clarity.

measurements indicate that the plasma is in a state of thermal non-equilibrium, with gas temperatures near room temperature and vibrational temperatures an order of magnitude higher. The high vibrational temperature relative to rotational temperature is common for ionizing plasmas.

Both T_{rot} and T_{vib} increase monotonically with increasing applied microwave field. This is due to the fact that electron-neutral collisions are responsible for excitation of both excited states and as the average electron energy is increased with the microwave field, so too is the transfer of energy to the molecular states. For fixed applied microwave field, both the measured vibrational and rotational temperatures are observed to decrease with increasing pressure. For the vibrational temperature, similar behavior has been observed at a frequency of 12 GHz,²⁴ where T_{vib} was found to decrease with increasing pressure to a minimum value near the minimum of the Paschen curve, which is near 30 Torr for this frequency. Our experiments show T_{vib} decreasing with increasing pressure up to 100 Torr, which is near the Paschen minimum for our frequency of 110 GHz. A theoretical model of the dependence of T_{vib} and T_{rot} on pressure would require knowledge of the plasma density as well as other plasma parameters which were not determined in these experiments. These measurements will be the subject of future research.

IV. CONCLUSION

Spectroscopic measurements of air breakdown initiated by a focused 110 GHz gyrotron beam have been carried out. Using the line-by-line radiation code SPECAIR, measurements of T_{rot} and T_{vib} have been obtained at pressures between 1 and 100 Torr and electric field strengths a few times greater than the threshold required for air breakdown. The molecular temperatures are found to be similar to DC and lower frequency breakdown experiments. The plasma is observed to be in a state of non-equilibrium, as vibrational temperatures are determined to be in the range of 4200–6200 K and rotational temperatures are in the range of 300–500 K. For this experiment, gas heating is found to be small as

rotational temperatures are not raised significantly above room temperature. Both rotational and vibrational temperatures are shown to increase with applied field and also depend on gas pressure.

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