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The role of translational and vibrational energy in processing plasmas: novel optical diagnostics of low-pressure Cl_2 and O_2 inductively-coupled plasmas

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A common assumption for "Low-temperature" plasmas is that neutral molecules and atoms in the system are in thermal equilibrium with the surrounding ambient (room) temperature, and only charged particles, which can acquire energy from applied electric fields, have higher mean energies. In reality, energy can be transferred from electrons or ions to the neutral gas, increasing the gas translational temperature. Furthermore, non-equilibrium vibrational or rotational distributions can occur in molecular gas plasmas. This can have significant effects on the plasma dynamics. Firstly, since most plasma reactors operate in a pressure-controlled regime, high gas temperatures will cause a considerable decrease in gas density (and therefore in electron-neutral collision rates). Secondly, the rates of activated processes may be significantly increased by translational energy.

We have developed a new, unambiguous technique to measure gas translational temperature of atoms, using Doppler-resolution Two-Photon Absorption Laser Induced Fluorescence (HR-TALIF) employing a specially-built narrow-bandwidth tuneable pulsed UV laser[1]. Results have been obtained on oxygen atoms, where a measurement precision of ±10K is readily obtained. In a DC glow discharge in pure O_2 the gas temperature up to 550K are observed. We have also applied this technique to atmospheric-pressure plasma jets, allowing the pressure-broadening coefficient to be determined[2]. The technique will be extended to the study of lower-pressure inductively-coupled plasmas, where higher temperatures are expected, and to chlorine atoms. This technique should also allow the determination of temperature profiles, from which the surface thermal accommodation coefficient can be derived. This key parameter has a strong impact on temperature modeling and is very poorly known[3].

Low-energy electrons can interact with molecules via resonances to cause vibrational excitation with large cross-sections. Such processes can absorb significant energy from the plasma electrons, affecting the electron energy distribution and potentially (via vibration-translation (VT) energy transfer) causing substantial gas heating. The presence of vibrationally excited molecules may significant increase the rates of collisional processes, including electron dissociative attachment and electron impact dissociation into neutral atoms. However, the cross-sections of these processes are often poorly known since they are extremely difficult to measure directly, and reliable theoretical calculations are only now appearing for simple diatomic molecules. We have developed a high-sensitivity ultra-broadband ultraviolet absorption spectrometer to measure vibrational distributions. This employs a highly-stable laser-plasma light source and achromatic optics, allowing absorption spectra over a 250nm range to be measured with a baseline stability of the order 10^{-5}. In pure O_2 discharges (both DC glow and in a low-pressure ICP reactor) we were able to observe oxygen molecules in vibrationally-excited levels up to v=18 (more than half-way to dissociation), with a "tail" vibrational temperature of 7000K [4]. Vibrational excitation was also detected in Cl_2 molecules in a pure Cl_2 ICP[5]. However, Cl_2 appears to be close to thermal equilibrium with the gas translational temperature, which nevertheless approaches 2000K in this case. We are developing detailed self-consistent 0D global models of these systems including vibrational excitation.

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