Editorial for plasma diagnostics using spectroscopic methods

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EDITORIAL

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Editorial

Editorial for plasma diagnostics using spectroscopic methods

Guest Editors

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Low-temperature plasmas have been used in various technological fields such as electronics, surface coating/modification, environmental engineering, medicine, agriculture, etc. While it is appreciated that the functional properties of plasmas are governed by the effects of electrons, ions, reactive neutral species, and electric fields, it remains a challenge to obtain detailed information about physical parameters in plasmas.

Much effort has been paid to the development of diagnostic methods which can be utilized for measuring electron density, electron temperature, densities of reactive neutral species, gas temperature, and electric field in low-temperature reactive plasmas. While many diagnostic techniques are based on intrusive methods (electrical probes, mass spectroscopy) that can nominally perturb the plasma state, continued effort focuses on developing non-intrusive techniques that primarily use optical signals to assess the plasma state.

This special issue highlights the most recent progress in optical techniques for measuring physical parameters in low-temperature reactive plasmas. The scope includes both passive (optical emission spectroscopy) and active (mainly laser-based spectroscopy) diagnostic methods. While historically, plasmas produced in gases at reduced pressures have been the focus of the low-temperature plasma community, atmospheric-pressure plasmas have recently attracted more and more attention. In line with this research trend, most papers in this special issue are related to the diagnostics of atmospheric-pressure plasmas.

Diagnostics of low-pressure reactive plasmas

Low-pressure (<1 Torr) reactive plasmas are used for fabrications of microelectronics devices and liquid crystal display panels. At the initial stage of the development of plasma-based technologies for electronics devices (approximately 30 years ago), engineers in industry had serious difficulty in the lack of understanding of radical densities in the plasmas they used for their material processing. Various optical methods, such as emission spectroscopy, absorption spectroscopy, and laser-induced fluorescence spectroscopy, have been developed for measuring the densities of reactive species in plasmas, and nowadays it has become possible to obtain detailed information on radical densities in low-pressure reactive plasmas. However, the measurement of electric fields in plasma is still difficult, and until recently the methods developed need expensive laser sources and sophisticated experimental skills. Nishiyama *et al* report on a new method for measuring sheath electric fields in hydrogen plasma [1]. They apply saturation spectroscopy to obtain the Stark spectrum of the Balmer-alpha line of atomic hydrogen, and deduce the sheath electric field with a detection limit of 10 V cm^{-1} . Since the light source is a diode laser, the system is compact and can be used in many experiments where knowledge of the sheath electric field is required.

Optical emission spectroscopy

Optical emission spectroscopy is an economical plasma diagnostic method. It is based on the recording of the spontaneous emission from a plasma. The spectra can consist of atomic line radiation, broadband recombination radiation, or molecular band emissions, to just name a few. The parameter which can be deduced from emission of a diatomic molecule is the rotational temperature which is believed to be equal to the translational temperature of the molecule in a collisional plasma environment. However, in atmospheric-pressure plasmas, we sometimes observe an optical emission spectrum which cannot be fitted by a

Boltzmann distribution corresponding to a rotational temperature. To solve the problem in the spectral fitting, Vorac *et al* developed a state-by-state fitting method and applied it to OH spectra observed in a surface barrier discharge [2].

The absolute line intensity observed by optical emission spectroscopy gives us the absolute density of the upper energy state of the transition line. In many cases, we are not interested in the absolute density of the electronic excited state of reactive species, since almost all the reactive species are in the ground state. However, when we detect the optical emission intensity of a forbidden line, we can obtain the density of the metastable (upper) energy state which can play an important role in processing and discharge physics. Inoue and Ono report on measurements of the density of molecular oxygen in the metastable singlet delta state by observing the optical emission intensity of the forbidden $a^1\Delta_g - X^3\Sigma_g^-$ line in atmospheric-pressure helium-oxygen plasma jet [3].

Densities of reactive species

Knowing the densities of reactive species is of utmost importance in applications of reactive plasmas. And by measuring the spatiotemporal variations of the densities we can e.g. obtain fundamental understanding on the kinetics of reactive species. Simek *et al* measured the evolution of the density of $N_2(A^3\Sigma_u^+, v = 0-8)$ in medium pressure (50 Torr) streamer discharge by laser-induced fluorescence spectroscopy, and discuss the influence of oxygen admixture on the formation of low vibrational levels [4]. The vibrational relaxation of $N_2(A^3\Sigma_u^+)$ is clarified by the elaborate experiment with detection of vibrational excited states up to $v = 8$. Another method to detect reactive species is absorption spectroscopy. Simeni *et al* report on the measurement of NO density and temperature in open-air confined plasma by mid-infrared absorption spectroscopy using a quantum cascade laser as the light source [5]. They achieve spatial resolution by applying Abel inversion to line-of-sight integrated absorption data.

The lifetime of an electronic excited state is not determined by the transition probability in atmospheric-pressure plasmas. This is because the frequency of collisional quenching exceeds the transition probability in the highly collisional environment. The collisional quenching causes a serious problem in the measurement of the density of reactive species. Martini *et al* investigated rate coefficients of collisional quenching and vibrational relaxation of $OH(A^2\Sigma^+, v = 0,1)$ in various gases [6]. Their data will be utilized by many researchers for estimating the density of $OH(A^2\Sigma^+)$ from relative emission intensities recorded during laser-induced fluorescence experiments.

Electron density and electric field

A measurement of the electron density in atmospheric-pressure plasma is a difficult task. Electrical probes and microwave (or optical) interferometry do not work at atmospheric pressure. Laser Thomson scattering is applicable, but sophisticated experimental skills are necessary to detect the weak scattered signal and to eliminate the stray light. The measurement of Stark broadening width is a simple method, but it is limited in a high-density plasma if a normal-size spectrograph is used in the experiment. Inada *et al* has adopted a Shack–Hartmann-type laser wavefront sensor to a streamer discharge and obtained 2D distributions of electron densities at a specific delay time after the trigger [7].

The measurement of electric fields is also challenging in atmospheric-pressure plasmas. Conventional Stark spectroscopy has difficulty in atmospheric-pressure plasmas because of the collisional quenching or the wide collisional broadening width. Simeni *et al* report electric field measurements in nanosecond pulsed discharges in atmospheric air by picosecond four-wave mixing [8]. This is a coherent anti-Stokes Raman scattering based method, but a tunable laser is not necessary in their system. They measure rapid variations of the electric field with a sub-nanosecond temporal variation.

Diagnostics of plasma-liquid interface

Plasmas in contact with solutions have attracted much interest very recently, in conjunction with the applications to medicine, agriculture, and environmental technologies. However, we have little knowledge on the plasma-liquid interaction at the moment. Kondo *et al* apply vibrational sum-frequency generation spectroscopy to water surface interacting with the spatial afterglow of a dielectric barrier discharge [9]. By this challenging attempt, they observe the change in the structure of the top surface of water.

We would like to thank all contributors to this special issue. We also hope that the diagnostic techniques reported in this special issue are utilized in experiments of many researchers and contribute to the progress in understanding of the physics and chemistry of low-temperature reactive plasmas.

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