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Metastable argon atoms under significant neutral depletion in helicon plasmas by laser-induced fluorescence

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ABSTRACT

We study the spatial distribution of the metastable-state argon atoms in high density helicon plasmas by means of laser-induced fluorescence. It is observed that the neutral argon in metastable-state has an anomalous radial distribution in density; it has a caldera-like shape radially, which is rare in typical low-temperature plasmas such as inductively coupled plasmas wherein the density increases toward the discharge center, as previously reported. The formation of the distribution can be explained as it forms by the combined effects of significant neutral depletion in high plasma density, off-axis electron-density distribution, and increasing diffusive loss toward the wall. To establish the assertion with the underlying physics, we calculate a simple global model and obtain the neutral density distribution in metastable-state under various conditions. The calculated results qualitatively agree with the experimental results.

1. Introduction

To date, the metastable state, which state is an energy level from which electric dipole radiation is forbidden, has been of an interest in various plasma-related fields because the metastable state has unusual properties such as a considerably longer life time. Further, it plays the role of an energy reservoir and exhibits anomalous evolutions with electron density and pressure [1–13]. In particular, in low-pressure and high-density plasmas, which are indispensable for the modern semiconductor and display industries, neutrals in the metastable state plays a crucial role in the discharge properties because they exist in a large fraction of ground-state atoms, and metastable-state reactions such as Penning ionization and metastable pooling frequently occur [2]. Moreover, it has been shown that neutral atoms in the metastable play a determining role in the kinetics of the discharges so that stepwise processes through the metastable state are of importance [9]. Therefore, the study of the properties of neutral atoms in the metastable state has become important in the context of the high-density plasma industry.

The helicon discharge is one of the interesting high-density plasma sources in low temperature glow discharge together with inductively and capacitively coupled plasmas (ICPs and CCPs, respectively); it is considered as one of the promising plasma sources for industry and possesses unusual physical properties such as abnormally high electron density at low neutral pressure, significant neutral depletion, and formation of a radially off-axis electron density distribution depending on the experimental conditions [14–17]. Even the mechanism of the helicon discharge is still disputed [18]. A study of neutral atoms in the metastable state in the helicon plasma is very important to understand its reaction in the utilization of helicon plasma for industry. However, the metastable-state species in helicon plasma has not been studied in detail due to complexity of measurement; on the other hand, the properties of the metastable state in both ICPs and CCPs have been clearly understood [1–3]. B Clarenchach et al. have reported on helicon discharge achieved by means of the time-dependent measurement of laser absorption [19]. The result, though significant, were limited to the antenna region. In the backdrop of the few studies on helicon discharge, the properties of the metastable state in helicon plasmas require a fuller investigation.

In this work, we study the metastable argon atoms in helicon plasmas by means of laser-induced fluorescence (LIF). To understand the underlying physics, we calculate a simple global model, which has been numerically investigated and experimentally confirmed in the ICPs [1–3]. For the calculation of the global model in helicon plasmas, the effect of neutral depletion, off-axis electron

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density distribution, and spatially varying diffusive loss are considered as the underlying physics.

2. Experimental details

Fig. 1 shows the schematic of the experimental setup of the ICP and helicon discharge [20]. A cylindrical Pyrex tube with inner diameter and height of 110 *mm* and 700 *mm*, respectively, is connected in series with two cylindrical stainless chamber: one chamber (B chamber, inner diameter and height of 110 *mm* and 140 *mm*, respectively) is utilized for both the laser path and signal detection while the other (inner diameter and height of 300 *mm* and 310 *mm*, respectively) is utilized as a diffusion chamber. All measurements were performed at the B chamber, which lies downstream of the discharge region. The driving antenna is a single-loop helicon antenna for $m = 0$, and the discharge is ignited and sustained by a 13.56 *MHz* rf current delivered by an RF power supply through a coaxial cable and L-type matching network (*Path Finder, PLASMART Inc.*). A uniform dc magnetic field of up to 600 *G* is applied with use of solenoid coils. The background base pressure in the vacuum chamber is in the range of 1×10^{-5} Torr and the operating pressure is in the range of 1×10^{-2} *Torr*. This operating gas pressure was maintained during the discharge operation via the circulation of argon gas (20 *sccm*) through the discharge chamber.

For the laser-induced fluorescence experiment, an optical parametric amplifier (OPA, Sunlite *Continuum Inc.*) was employed [2,3]. A frequency-tripled (third-harmonic generation of 1064 *nm*) Nd:YAG laser (Powerlite 9010, *Continuum Inc.*) working at 10 Hz with a pulse energy of 300 *mJ* at 355 *nm* was used as the pumping laser of the OPA. The OPA used in the study has a broadband spectrum with a full width at half maximum of about 5 *nm* when compared with the conventional dye laser, which is normally used for LIF; consequently, we utilized a fine-resolution spectrometer because the width of the LIF signal was considerably less than 1 nm [21]. As regards the LIF signal acquired to measure the metastable-state density, the $1s_5$ state was pumped by the laser at 696.5 *nm* to the $2p_2$ state of the 4p excited states, and spontaneous emission at 696.5 *nm* from the $2p_2$ state to the $1s_5$ state was detected. Although the laser wavelength of both the incidence and detection beams was the same, these beams could be easily distinguished because the bandwidth of the incident laser was considerably broader than the detection wavelength [2].

The LIF signals were collected by two lenses and a 10 *m* optical fiber bundle, since two lenses positioned in front of an optical fiber can be used to measure signals from a certain measurement position (the focusing point). The horizontally polarized laser beam parallel to the external magnetic field to provide absorption of only one π Zeeman component of the line [19] rotated by a $\lambda/2$ waveplate having broadband spectral width (690 *nm* ~ 1200 *nm*) was focused using a lens (focal length $f = 1000$ *mm*) at the point of measurement, located downstream of discharge. For radially spatial measurements of LIF, we installed a translational stage behind the laser focusing lens and collecting optics, which is composed of two lenses and an optical fiber on an optical board so that movement of a measured point could be made possible. For this measurement, precise alignments of the optics (lens and fiber) are required, and therefore, we used a diode laser for alignment. An intensified charged coupled device (ICCD) camera was affixed to one side of the monochromator, which camera operates with doubled trigger signals (20 *Hz*) generated from a laser trigger (10 *Hz*) to simultaneously extract background signals emitted by the plasma. The detected LIF and background emission signals were recorded and transferred to a computer for further data processing.

Fig. 2 shows the measured LIF spectrum along with stray laser light. As shown, the measured spectrum includes the stray laser light (black line) that is reflected from the chamber wall and windows along with LIF signals because we used the same wavelength of the laser spectrum with LIF spectrum centered at 696.5 *nm* in both cases. The Rayleigh Scattering (RS) signal can be neglected because the laser energy is too weak to detect RS signals. By subtracting the stray laser light and background noise, we could obtain a pure LIF spectrum.

3. Results and discussion

In the discharge, one of three different modes, named the E (capacitive) mode, H (inductive) mode, and W (wave) mode depending on the rf power and strength of the applied dc magnetic (B) field, can be sustained [22]. Among the modes, the W mode can easily be identified by the observed blue emission (named here as "blue core") when argon is formed along the magnetic field axis in a cylindrical hollow-core shape [15,16]. Under a relatively weak

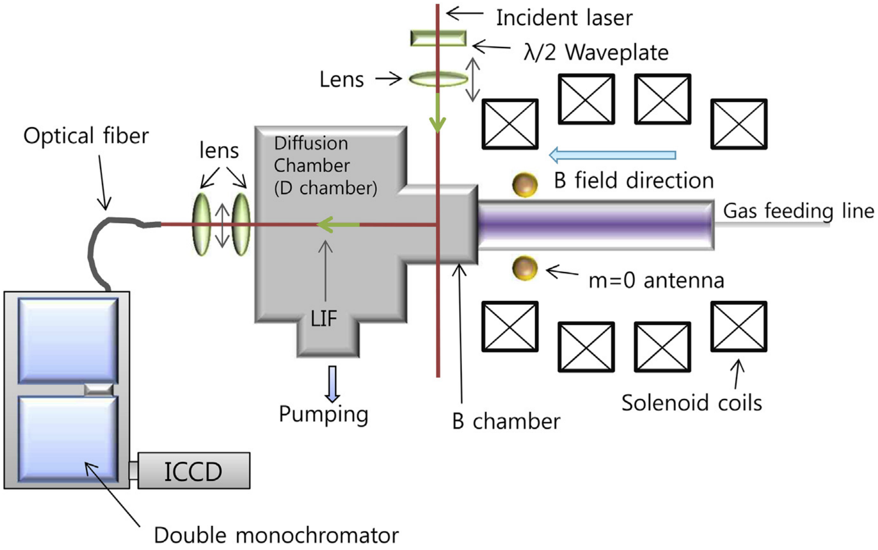

Fig. 1. Schematic of the experimental setup for laser-induced fluorescence experiment in helicon plasmas.