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ABSTRACT
The expanding cascaded arc Ar/N$_2$ plasma has been investigated by both the active and passive optical diagnostic technologies. In the investigation, the laser Thomson scattering (LTS) and optical emission spectroscopy (OES) have been adopted to measure electron temperature ($T_e$) and electron excitation temperature ($T_{\text{exc}}$), respectively. The LTS measurements show that a remarkable nonlinear behavior of $T_e$ as a function of the N$_2$/(Ar + N$_2$) ratio is found, which is caused by the collective interaction between the superelastic collision and the electron-impact excitation. The superelastic collisions by the highly excited vibrational nitrogen molecules can effectively heat the free electrons, while the electron kinetic energy can be transferred to N$_2$ molecular internal energy via the electron impact with N$_2$ in the ground state. The difference between $T_{\text{exc}}$ and $T_e$ demonstrates that the cascaded arc Ar/N$_2$ plasma significantly deviates from the local thermodynamic equilibrium. This would be useful for improving our further understanding of nonequilibrium plasma and extending applications of the cascaded arc Ar/N$_2$ plasma.

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I. INTRODUCTION
Cascaded arc plasma source can generate high electron density (up to $10^{22}$ m$^{-3}$), low electron temperature, and steady-state plasma and has widely been applied in many fields, such as chemical vapor deposition, etching, material surface modification, and linear plasma simulator. However, the physical mechanism of the cascaded arc discharge under the mixture gases conditions, especially for the reactive gases (such as N$_2$, O$_2$), is still not fully understood. Therefore, to interpret the physical processes in the plasma, it is crucial to directly diagnose the plasma parameters. In particular, the electron temperature ($T_e$) which relates to the electron kinetic energy is one of the most important physical parameters of plasma. Nevertheless, this concept seems to be confusing sometimes because different “temperatures” can be obtained when using different diagnostic approaches.

The laser Thomson scattering and the optical emission spectroscopy are two important and commonly used plasma diagnostic methods. As an active plasma diagnostic technology, LTS allows one to obtain $T_e$ accurately without any prior assumption on the state of departure from the equilibrium of plasma. This technique has been developed as one of the most accurate approaches and utilized to diagnose various types of plasmas. Compared with LTS, OES does not require a complicated experimental setup. It has widely been applied to measure the temperature as well. Although both LTS and OES can determine the plasma temperature ($T_p$), $T_p$ measured by LTS is the real electron temperature, namely, the electron kinetic temperature ($T_e$), while $T_p$ obtained by OES is known as the electron configuration temperature or electron excitation.
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temperature \( (T_{\text{exc}}) \). \( T_{\text{exc}} \) can be regarded as \( T_e \) only when the plasma is in the local thermodynamic equilibrium (LTE).\(^{12} \) If the plasma deviates from LTE, \( T_{\text{exc}} \) will not be equal to \( T_e \) any more. In this case, the difference between \( T_{\text{exc}} \) and \( T_e \) can be utilized to evaluate whether the plasma satisfies the condition of LTE.

In the cascaded arc plasmas, Zhou et al.\(^{22} \) and Meulenbroeks et al.\(^{23} \) reported that the expanding cascaded arc plasmas with H\(_2\) and Ar-H\(_2\) gases could deviate from LTE by measuring the atomic state distribution function, respectively. In this work, the cascaded arc plasma with Ar and N\(_2\) gases has been investigated by LTS and OES. We analyzed the deviation from LTE in Ar plasma with adding N\(_2\) gas via the comparison between the “electron temperatures” measured by these two approaches.

II. EXPERIMENTAL ARRANGEMENTS

A. Laser Thomson scattering diagnosis system

The laser Thomson scattering was employed to measure the electron temperature of the cascaded arc Ar and N\(_2\) mixture plasma. A schematic of the experimental setup is shown in Fig. 1. The experimental system consisted of several subsystems: plasma generator, laser probe system, LTS collecting system, and detecting system.

1. Plasma generator subsystem

The plasma was generated in a cascaded arc plasma source,\(^2,24 \) which mainly comprised of a needlelike tungsten cathode, three copper cascaded plates, and an anode plate. The plasma source was mounted on a flange connected to the vacuum chamber with an ultimate background pressure of 0.1 Pa. The plasma was injected into the vacuum chamber, and then, it expanded along the axial direction with the radial diffusion. In this work, Ar gas was the major gas and N\(_2\) gas was added as the trace gas. We analyzed the deviation from LTE in Ar plasma with adding N\(_2\) gas via the comparison between the “electron temperatures” measured by these two approaches.

2. LTS laser probe subsystem

As shown in Fig. 1, the investigations were performed by a 10 ns pulsed Nd:YAG laser with a wavelength of 532 nm at a repetition rate of 30 Hz. The maximum laser energy is 325 mJ. After being guided by four mirrors (M1-M4), the laser beam was focused into the detection volume by a planoconvex lens with a focal length of 1 m. The detection volume was about 150 mm away from the nozzle of the plasma source. To suppress the stray light, Brewster windows were mounted on the laser input and output arms in which several baffles were placed. In order to monitor the laser power pulse-to-pulse, a fraction of laser beam was picked off by a beam sampler to the photodetector which was protected by a neutral density filter.

3. LTS collecting subsystem

The Thomson scattering light was perpendicularly collected in this work. A planoconvex lens with a focal length of 150 mm focused the scattering light onto a fiber bundle constituting 19 separated fibers in a linear array (19 \( \times \) 1). The collecting subsystem was optically shielded from the external environment to prevent the stray light from entering into the fibers. Besides, a viewing dump was adopted to further reduce the stray light.

4. LTS detecting subsystem

The scattering light was transferred to the detecting subsystem by the fiber bundle with the length of 10 m. The detecting subsystem was composed of a homemade Triple Grating Spectrometer (TGS) and a high quantum efficiency iCCD camera. The TGS was considered as a spectrometer combining a notch filter, and it can greatly suppress Rayleigh scattering and stray light. This optical configuration has been reported in other laboratories.\(^{25–29} \) An iCCD camera was utilized to detect the Thomson scattering signal and record the Thomson scattering spectra. In addition, a delay generator (DG645) was employed to synchronize the laser and iCCD camera.

In this work, the Thomson scattering spectra were obtained by integrating multiple laser pulses and the gate width of the iCCD was

FIG. 1. The schematic diagram of laser Thomson scattering diagnosis for the cascaded arc Ar plasma with the N\(_2\) addition. M1-M4: mirrors; A1, A2: apertures. A typical Thomson scattering spectrum is illustrated in the upper-right corner.
FIG. 2. The schematic diagram of OES for measuring the electron excitation temperature of the plasma.

set as 20 ns. The pixels of iCCD in the vertical direction were binned to achieve a good signal to noise ratio value of LTS signal at the sacrifice of the spatial resolution. A typical Thomson scattering spectrum is shown in the insert of Fig. 1.

B. Optical emission spectroscopy diagnosis system

As shown in Fig. 2, the collecting subsystem of OES was identical to that of LTS. When the OES measurement was carried out, the head of fiber bundle connected to TGS in the LTS experiment was switched to a commercial spectrometer (Andor Shamrock 750) which has been calibrated by a broadband light source (labsphere, LPS-400). The OES light was dispersed by the spectrometer, and the spectra were detected and recorded by using an iCCD camera. The discharge conditions in OES measurements were set as the same as those in the LTS experiments. A typical cascaded arc Ar plasma spectrum is presented in Fig. 3(a), and six Ar atomic emission lines (675.28 nm, 687.13 nm, 703.03 nm, 706.72 nm, 727.29 nm, and 750.39 nm) were chosen to calculate $T_{\text{exc}}$ via Boltzmann plot, as shown in Fig. 3(b).

III. RESULTS AND DISCUSSION

A. The electron temperature as a function of $N_2$ addition ratio

Figures 4(a) and 4(b) show $T_e$ as functions of $N_2$ ratio from 0% to 10% and discharge currents from 80 A to 120 A at the background pressure of 500 Pa and 800 Pa, respectively. The results indicate that $T_e$ grows first to a maximum and then decreases with the increase in the $N_2$ content. Similar behaviors have been observed at the low pressure of 150 Pa and 300 Pa.

Figures 5(a) and 5(b) present that maximum $T_e$, i.e., the “turning point” of $T_e$ illustrated by the dashed-dotted lines in Fig. 4(a), appears in the higher $N_2$ ratio with the enhancement of discharge current.

For the expanding Ar cascade arc plasma, van de Sanden et al. have demonstrated that the three-body recombination reaction (1) plays an important role in the electrons heating,

$$Ar^+ + e + e \rightarrow Ar + e.$$  

However, when the $N_2$ gas is fed into the Ar plasma, both the Ar ion density ($n_{\text{Ar}^+}$) and electron density ($n_e$) drastically drop due to the charge exchange reaction (2) and the dissociative recombination reaction (3).31,32 Both the reactions (2) and (3) are the dominant processes in the expanding plasma containing molecules.33,34 Hence, the three-body recombination contribution becomes gradually weak when more and more $N_2$ gases are fed into the plasma;

$$Ar^+ + N_2(X, v) \rightarrow Ar + N_2^+(X, v),$$

$$N_2^+(X, v = 0, 1) + e \rightarrow N(S) + N(D).$$

Furthermore, van de Sanden also verified that the Ohmic dissipation of the generated current density in combination with the electron heat conduction can preheat the electrons in the shock front. However, no shockwave is observed in our plasma so that the electrons are hardly heated by this mechanism.

FIG. 3. (a) A typical spectrum of the cascaded arc Ar and $N_2$ admixture plasma and (b) the Boltzmann plot of the Ar I lines marked. The H$_\alpha$ line in (a) was from the residual water vapor in the vacuum chamber.
FIG. 4. Electron temperature measured by LTS as a function of \(N_2\) concentrations with different discharge currents at 500 Pa (a) and 800 Pa (b), respectively. The dashed-dotted lines in (a) show the \(N_2\) ratio where maximum \(T_e\) appears.

FIG. 5. The correlation between the \(N_2\) ratio corresponding to maximum \(T_e\) and the discharge current at 500 Pa (a) and 800 Pa (b), respectively.
FIG. 6. Comparison of $T_\text{e}$ measured by LTS (black) and $T\text{exc}$ measured by OES (red) in the different discharge currents at 150 Pa (a), 300 Pa (b), 500 Pa (c), and 800 Pa (d), respectively.
The highly excited vibrational ground state \([N_2(X, v^*)]\) can transfer the energy to free electron very effectively in a rather short nanosecond-time scale.\(^{36-39}\) This is called superelastic collision. Therefore, the superelastic collision between \(N_2(X, v^*)\) and free electrons can effectively heat the electrons in an expanding cascade arc plasma. The species of \(N_2(X, v^*)\) are produced via the following reaction:

\[
N(S, P, D) + N_2(X, v = 0) \rightarrow N_2(X, v^*) + N. \tag{4}
\]

The \(N\) atoms in reaction (4) are generated via the dissociative recombination reaction. Due to the full dissociation of molecules in the arc channel,\(^{36-38}\) the \(N_2\) molecules in reaction (4) are formed at the vessel wall following the diffusion of \(N\) atoms and then reenter to the detection volume. As a result, \(T_e\) rises with the addition of \(N_2\) gas via the superelastic collision,

\[
N_2(X, v^*) + e \rightarrow N_2(X, w) + e. \tag{5}
\]

However, when more \(N_2\) gases are fed into the Ar plasma, both \(n_{Ar^+}\) and \(n_e\) become too little to produce the adequate energetic \(N\) atoms which leads to an insufficient production of \(N_2(X, v^*)\). In this case, the electrons cannot be heated effectively via the superelastic collision. Meanwhile, the increase in the ground state \(N_2\) molecules' \([N_2(X, v = 0)]\) density enhances the electron-impact excitation process which induces that electrons lose more kinetic energy.

\[
e + N_2(X, v = 0) \rightarrow e + N_2(X, v = 1, 2, \ldots). \tag{6}
\]

Therefore, in the higher \(N_2/(Ar + N_2)\) ratio, \(T_e\) declines due to the weak superelastic collision heating and the enhanced electron energy loss.

When the discharge current increases, more input power is injected into the plasma source. Hence, both \(n_{Ar^+}\) and \(n_e\) increase. The species of \(N_2(X, v^*)\) via reaction (4) following reactions (2) and (3) are sufficient to heat the electrons by the superelastic collision. As a result, the "turning point" of \(T_e\) shifts to a higher \(N_2/(Ar + N_2)\) ratio value with increasing the discharge current, as shown in Figs. 5(a) and 5(b).

**B. Deviation of \(T_{exc}\) from \(T_e\) as a function of \(N_2/Ar\) ratio**

The correlations between \(T_e\) (electron kinetic temperature) and \(T_{exc}\) (electron excitation temperature) in different discharge conditions are shown in Figs. 6(a)–6(d). It can be clearly seen that there is a significant discrepancy between \(T_{exc}\) and \(T_e\). The values of \(T_{exc}\) are always greater than those of \(T_e\) in the given plasma conditions. The influence of \(N_2\) gas on \(T_{exc}\) is also different from that on \(T_e\). In order to evaluate the difference between \(T_{exc}\) and \(T_e\), the relative error, which is defined by \([T_{exc} - T_e]/T_e \times 100\%\), is introduced and shown in Fig. 7. The results indicate that the addition of \(N_2\) leads to a significant enlargement of the relative errors, even up to 100%.

If the plasma deviates from LTE condition, \(T_{exc}\) will not be equal to \(T_e\), which indicates that \(T_{exc}\) does not directly relate to the electron kinetic energy.\(^{22,40}\) Nevertheless, the comparison between \(T_e\) and \(T_{exc}\) provides a way to estimate the equilibrium deviation of plasma.\(^{12}\) As shown in Figs. 6 and 7, these differences between \(T_{exc}\) and \(T_e\) imply that the plasma is in the state of non-LTE.

When the plasma is in LTE, the populations in the different discrete quantum levels and continuous level are distributed according to the Saha-Boltzmann equilibrium as shown in Fig. 8(b). These are defined as Saha’s configuration values. If the populations in the levels exceed Saha’s configuration values as shown in Fig. 8(a), the plasma refers to the ionizing plasma.\(^{24,42}\) When the levels are underpopulated with respect to Saha’s configuration values which are presented in Fig. 8(c), the plasma belongs to the recombining plasma.\(^{41,42}\)

There is no external power to heat plasma in the downstream expanding zone of the cascaded arc plasma. Therefore, the recombination process dominates in the plasma. Meulenbroeks\(^{22}\) and Benoy\(^{41}\) have measured the values of \(b(p)\) which is expressed by Eq. (7) with the different quantum levels \(p\) in the expanding cascaded arc plasma,

\[
b(p) = n(p)/n_s(p), \tag{7}
\]

where \(n(p)\) is the real populations of the quantum level \(p\) and \(n_s(p)\) is the theoretical Saha’s configuration values of this level. The results show that the \(b(p)\)-factor is much smaller than 1. The plasma is in a nonequilibrium recombination phase. This demonstrates that plasmas in the present experiments belong to the recombining plasma.

In the recombining plasma, \(T_{exc}\) is always greater than \(T_e\).\(^{24}\)

In Fig. 8(c), the downward recombination flow from the continuous state (ion phase) to the ground state is dominant in the Ar atomic excitation space, which means that in the recombining plasma, the populations of Ar quantum levels are mainly produced by the radiative recombination from the continuous state of \(Ar^*\) and electron-impact de-excitation from the higher levels of Ar atoms.\(^{31,43}\) Both the radiative recombination and electron-impact de-excitation processes are predominated by the free electrons. However, when the \(N_2\) gas is fed into the plasma, the electron density drops rapidly due to the dissociative recombination reaction,\(^{24}\) which causes that both the radiative recombination and electron-impact de-excitation are weakened. The productions of \(Ar\) quantum excited levels’ population decrease, and the originally underpopulated levels further deviate from Saha’s configuration values. Therefore, the addition of
FIG. 8. The schematic of quantum level populations in the plasma in different phases. (a) Ionizing plasma. (b) Plasma in LTE. (c) Recombining plasma. The arrows represent the upward ionizing/downward recombining flow in the atomic excitation phase.

N₂ results in that the plasma significantly departs from LTE. This is the reason why the observed $T_{\text{exc}}$ deviates from the measured $T_e$ with the introduction of N₂ gas.

IV. CONCLUSION

In summary, both the laser Thomson scattering and optical emission spectroscopy have been developed to diagnose cascaded arc Ar/N₂ plasma. The results show that the N₂ gas has a significant influence on $T_e$. The superelastic collision between the free electrons and the highly excited vibrational nitrogen molecules can heat the free electrons to increase $T_e$, while the electron-impact excitation with the nitrogen molecules in the ground state can transfer the electron kinetic energy to the nitrogen molecule internal energy, leading to the reduction in $T_e$ at higher N₂ concentration. These two mechanisms collectively induce that the electron temperature $T_e$ has a nonlinear and nonmonotonic behavior as a function of N₂/(Ar + N₂) ratio. The N₂ gas also results in that the observed electron excitation temperature ($T_{\text{exc}}$) seriously departs from the real electron temperature. This investigation provides an approach to evaluate the deviation of LTE in the reactive Ar/N₂ plasma via comparing $T_{\text{exc}}$ with $T_e$. This would be useful for extensive applications of the other nonequilibrium plasma sources.

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