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Spectroscopic Study of a Boron-Nitride Capillary-Discharge Plasma

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Plasma radiation from a boron-nitride (BN) capillary discharge with an input energy density of ∼2 kJ/cm^3 has been analyzed using a 2-m grazing-incidence spectrograph. The electron temperature and the electron density of the plasma were obtained from the recombination continuum slope beyond the Lyman series limit of the B^{4+} ion and from the Stark broadening of the B V Lyman δ line, respectively. The time-averaged electron temperature and density thus measured were approximately 30 eV and 4×10^{19}/cm^3, respectively. However, the presence of Ca XII lines indicated that the peak electron temperature was as high as 150 eV. The spectroscopic data showed an intensity inversion between the L_β and the L_γ lines of the B^{4+} ion. Possible causes of the intensity anomaly are discussed.

I. INTRODUCTION

One of the current interests in laser physics is the generation of a table-top soft-X-ray laser. Because of its better efficiency and easy handling relative to the conventional soft-X-ray lasers which are usually driven by powerful optical lasers, the possibility of obtaining soft-X-ray amplification using capillary discharge plasmas has been studied during past years [1–4]. Recently, soft-X-ray amplification based on the recombination-pumping scheme in the C VI Balmer alpha (H_α) transition has been observed using a polyethylene capillary [5]. The result encouraged us to investigate the possibility of obtaining lasing action in other atoms. Since boron nitride (BN) is a good insulator and the atomic numbers of boron and nitrogen are next to carbon, a BN composite capillary might be a good material to be studied within the discharge parameters used in the polyethylene ($(CH_2)_n$) capillary.

In this paper, we present a study of a BN capillary discharge plasma through time-integrated and space-resolved spectroscopy in the wavelength range of 2.5 to 27 nm. The spectra showed strong B IV and B V spectral lines and many impurity lines. The electron temperature and density for the boron plasma were estimated to be T_e∼30 eV and n_e∼4×10^{19}/cm^3, respectively. It was found that there is an intensity inversion between the n=4 and 3 states. However, it is not likely that B V H_α lasing based on the three-body recombination scheme takes place, judging from the measured plasma parameters. The intensity inversion could be caused by either an opacity effect or a population inversion due to resonant charge-transfer between B^{5+} ions and neutral atoms.

II. EXPERIMENTAL SETUP

A schematic diagram of a capillary discharge system is shown in Fig. 1(a). The energy stored in two capacitors (C=60 nF each) was released through drilled carbon electrodes to a BN [6] capillary when a trigger pulse was applied. The trigger electrode [indicated as F in Fig. 1(a)] was made of carbon and was placed near one of the electrodes located opposite the XUV spectrograph. Copper sheets were used as a transmission line between the capacitors and the capillary. Capillaries of 1.05 to 1.3 mm in bore diameter and 8∼16 mm in length (D in the figure) were used. The capillary was evacuated to a vacuum pressure in the range of 10^{-4} to 10^{-5} Torr.

Each discharge was monitored by a Rogowski coil. An oscillogram of the current waveform of a 14-mm-long BN capillary discharge is shown in Fig. 1(b). The maximum current was 21 kA at a charging voltage of 25 kV, and the half cycle of the discharge was 220 nsec. The oscillogram shows a nearly critically damped harmonic oscillation. This corresponds to an inductance of 36 nH

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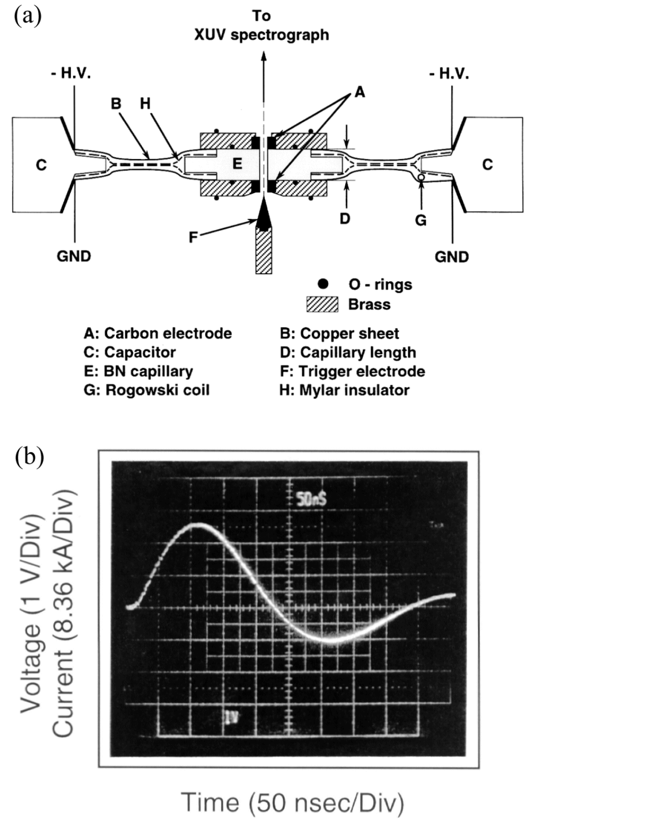

Fig. 1. (a) A schematic diagram of the capillary discharge system. (b) An oscillogram of the current waveform from a 14-mm-long capillary discharge for an input energy density of 2 kJ/cm^3 at charging voltage of 25 kV.

and a resistance of 0.3 ohm during the 1st half cycle of the discharge. Compared with the polyethylene capillary discharge [5], the BN discharge plasma has a larger resistivity. The BN capillary was completely destroyed after 5 to 10 discharges. Small craters and cracks developed in the capillary wall after a few discharges, while in the case of polyethylene the wall was smoothly enlarged due to a uniform ablation.

Using a toroidal mirror, soft X-rays emitted from the BN capillary discharge plasma were collected into a 2-meter grazing-incidence spectrograph through an entrance slit of 6 μm in width. A 600-lines/mm grating with an incidence angle of 1.5 degree was used. The spectra were recorded on KODAK 101-05 plates and scanned by a densitometer, and the photographic densities were transformed to spectral intensities by Henke's formula [7]. The toroidal mirror increased the recording speed and served to obtain space-resolved spectra [5,8]. Optical parameters were set such that the stigmatic wavelength (focus) on the meridional plane was 28.7 nm. In the sagittal plane, the toroidal mirror formed an astigmatic image of the source at the entrance slit.

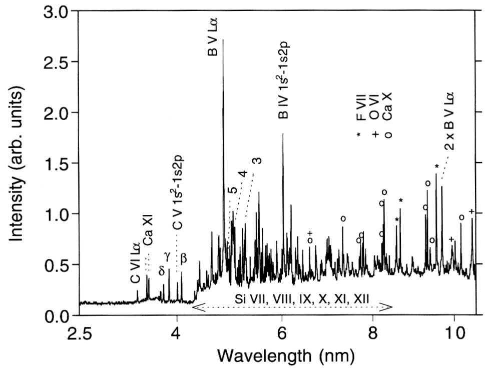

Fig. 2. A time-integrated spectrum from a 14-mm-long capillary plasma in the wavelength region of 2.5 to 10 nm. Fifteen discharge exposures using four new BN capillaries were recorded. The input energy density is 2±0.4 kJ/cm^3. The identified spectral lines are summarized in Table 1.

III. RESULTS

Figure 2 shows the time-integrated spectrum from a 14-mm BN capillary plasma, viewed on the capillary axis, with an input energy density of 2 kJ/cm^3 in the wavelength range of 2.5 to 10 nm. The energy density is defined as stored energy (CV^2/2) divided by the capillary volume. Fifteen discharge exposures using four new BN capillaries were made in order to obtain the spectrum. The reciprocal dispersion is 0.62 Å/mm at 4 nm and 0.94 Å/mm at 10 nm. The estimated instrumental width is 50 mÅ.

As shown in Fig. 2, impurity lines from $C^{4+,5+}$, O^{5+}, F^{6+}, $Si^{6+,7+,...,11^+}$, and $Ca^{9+,10+}$ ionic species, as well as the strong resonance lines of B V 1s-*n*p and B IV 1s^2-1s*n*p, dominate the spectrum. The identified lines are summarized in Table 1. The average charge state of a certain element can be obtained from the recorded spectrum, allowing one to infer the electron temperature [9]. The dominance of B V and F VII spectral lines suggests that the average temperature was in the range of 45 eV. On the other hand, the occurrence of Si VII,..., XI and Ca X, XI spectral lines indicates that the temperature was in the range of 50∼150 eV. The appearance of Ca XI spectral lines at 3.5 nm and Ca XII lines at 14.1 and 14.7 nm (see Fig. 5) and the abundant Si XI, XII lines in the wavelength range of 4.3 to 7.0 nm implies that the peak temperature was even higher. This indicates that the temperature at peak current is high enough to produce abundant fully stripped B^{5+} and C^{6+} ions. Figure 3 shows the detailed spectrum in the wavelength region of 3.0 to 4.2 nm. The intensity of the spectrum is normalized to the B V L_α peak (not shown) intensity for convenience.

The electron temperature for the boron plasma was obtained from the recombination continuum from the B^{5+} ion. In the B VI recombination continuum, im-

Table 1. Identified spectral lines for the BN capillary discharge.

Ionic Species	Ionization Energy (eV)	Configuration	Spectral Range (Å)
B IV	259.377	$1s^2 - 1snp$ $1s2l - 1snl$	48.94∼60.31, 344, 385, 418.6
B V	340.229	$1s - np$, $2l - 3l$	37∼48.59, 194.35, 262
C V	392.090	$1s^2 - 1s2p$	40.27
C VI	489.997	$1s - 2p$	33.74
O VI	138.188	$1s^2 2l - 1s^2 nl$	95.1∼173, 184
F VII	185.188	$1s^2 2l - 1s^2 nl$	74.5∼128, 134.9
Si VII	246.53	$2s^2 2p^4 - 2s^2 2p^3 nl$	59.97∼88
Si VIII	303.18	$2s^2 2p^3 - 2s^2 2p^2 nl$ $n = 3$, (4)	51.8∼72.3, (214∼217)
Si IX	351.11	$2s^2 2p^2$, $2s2p^3 - 2s2p^2 3l$, $2s^2 2p3l$ $l =$ s, d	51.1∼67, (225∼227)
Si X	401.38	$2s2p^2$, $2s^2 2p$, $2p^3 - 2p^2 3l$, $2s2p3l$, $2s^2 3l$ $l =$ s, p, d	44.85∼66.7
Si XI	476.08	$2s2p$, $2p^2$, $2s^2 - 2p3l$, $2s3l$ $l =$ s, p, d	43.29∼52.3
Si XII	523.52	$1s^2 2p$,(2s) $- 1s^2 3d$,(3p)	(40.95), 44.165
Ca X	211.28	$2p^6 3s$, 3p, 3d $- 2p^6 nl$ $n = 4, 5, 6, 7$	73∼167
Ca XI	591.9	$2s^2 2p^6 - 2s^2 2p^5 3s$	35.21, 35.58
Ca XII	657.2	$2s^2 2p^5 - 2s2p^6$	141.04, 147.28

*Weak lines are noted with ( ) notation.

purity lines are negligible, except for the conspicuous Ca XI lines. When a Boltzmann distribution is assumed, the intensity of the recombination continuum scales as $I \sim \lambda^{-2} \exp(-hc/kT_e\lambda)$ [10]. The plot of $\ln(I\lambda^2)$ as a function of $1/\lambda$ should be linear, with the slope yielding the electron temperature. The electron temperature thus measured for the plasma was about 30 eV. This value is in the range of a previous estimate obtained by using an average charge state with the table of Post *et al.* [9].

The electron density was estimated from the Stark broadening of the B V L_δ line. The spectral line broadening of the L_δ line arising from Z≥6 elements for a given electron temperature and electron density has been summarized in Ref. 11. In order to apply these data to B^{4+}, the Z^{-5} scaling law [11–13] was used. By extrapolating the temperature scale to the values for the boron plasma, the FWHM (full width at half maximum) of the Stark-broadened profile was obtained as a function of electron density. The profile of the Stark-broadened L_δ line is basically Lorentzian and can be extracted from the experimental Voigt profile. To obtain the instrumental width near the L_δ line, the Ca XI $2p^6$-$2p^5 3s$ line was used

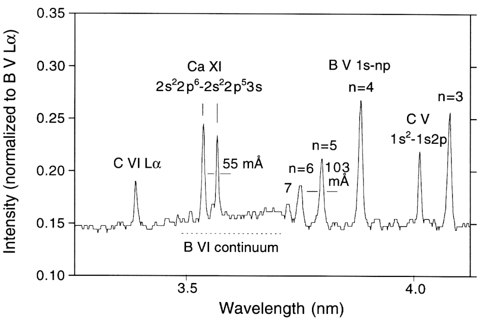

Fig. 3. The detailed spectrum of Fig. 2 in the wavelength region of 3.0 to 4.2 nm. For convenience, the intensities are normalized to that of the B V L_α line (not shown). The spectrum shows an intensity inversion between the L_β and the L_γ lines.

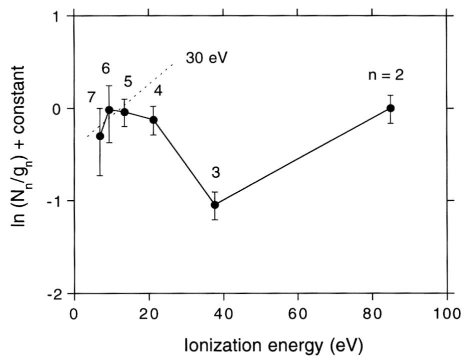

Fig. 4. Relative populations of B V states as a function of the ionization energy, showing a strong population inversion between the n=4 and 3 states.

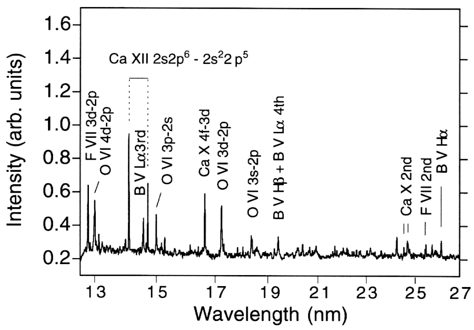

Fig. 5. Time-integrated spectrum from a 14-mm-long capillary plasma in the wavelength region of 13 to 27 nm.

since this line is most likely not Stark broadened, being a low-n transition line in a high charge-state ion. The measured line width was 55 mÅ, and this value is nearly the same as the calculated instrumental width of 50 mÅ for the XUV spectrograph. Since the Doppler broadening of the B V L_δ line is less than 5 mÅ for the 10 to 40 eV electron-temperature range, we set the Gaussian width of the spectral line as 55 mÅ. The measured FWHM of the L_δ line profile was 103 mÅ, and the estimated deconvolution of Lorentzian contribution resulted in an electron density of n_e=4×10^{19} cm^{-3} with an accuracy of 30%.

One feature of the spectrum obtained with the BN capillary discharge was an intensity inversion between the L_β and the L_γ lines, as can be seen in Fig. 2 and 3. This could be an indication of a population inversion between the n=4 and 3 states. The integrated spectral intensity for a given $n \rightarrow m$ transition is given by [14]

$$I \sim h\nu A_{mn} N_n \quad (1)$$

where A_{mn} is the spontaneous emission ($n \rightarrow m$) coefficient and N_n is the population of the upper state. The sublevel population N_n/g_n is then $I/(h\nu A_{mn} g_n)$, where g_n is the statistical weight. Assuming a local thermodynamic equilibrium (LTE),

$$N_n/g_n \sim \exp[(E_\infty - E_n)/kT_e] \, , \quad (2)$$

the logarithm of the sublevel population should give a positive slope as a function of the ionization energy $E_\infty - E_n$. However, the measured values for the n=2, 3, 4, and 5 states relative to the n=2 state were 1, 0.35, 0.88, and 0.96, respectively, with estimated errors of ±15%, showing that there may be a large population inversion between the n=4 and 3 states and between the n=5 and 3 states.

Figure 4 is a plot of the logarithm of the sublevel populations as a function of the ionization energy of each state. The plot clearly shows an apparent population inversion between the n=4 and 3 states. If this is the case, then a considerable amplification of the spontaneous emission (ASE) takes place along the capillary length at wavelengths in the vicinity of the 74.5-nm (n=4–3) transition line of the B^{4+} ion. The inversion factors $F = 1 - (N_n/g_n)/(N_m/g_m)$ for these transitions are in the range of 0.5 ∼0.7, representing a fairly high population inversion. However, the n=5 to 3 inversion is not likely to produce ASE because a considerable Stark broadening of this transition line greatly reduces its gain.

A similar intensity inversion were observed [15,16] in expanding laser-produced plasmas, which were initially generated by focusing a glass laser on various disc targets. The driving laser had an energy and a pulse width (FWHM) of 5 J and 10 ns, respectively. The intensity inversion was observed at some distance (typically 10∼20 mm) from the target. In that case, the intensity inversion was reported to be caused by the resonance charge-transfer recombination of B^{5+} (or C^{6+}) with neutral boron (or carbon). The electron density and temperature at a distance of 10 mm were inferred to be of the order of 10^{16} cm^{-3} and a few electron volts, respectively.

However, in the case of the BN capillary plasma, the electron density is about three orders of magnitude higher than in the expanding laser-plasmas described above, and the local thermodynamic equilibrium will be established rapidly (10^{-11} sec) for the $n \geq 3$ states, indicating that the population inversion will deteriorate within a very short period of time unless the n=3 state depopulates rapidly. However, the fast decay of the n=3 state can be impeded by radiation trapping when the lasant plasma is optically thick for the n=3 to 2 and 3 to 1 transitions. For a reasonable plasma depth (or diameter, say, 0.3 mm), the population inversion between the n=4 and 3 states can not be established in a boron-plasma with an electron density greater than 10^{18} cm^{-3} if it is only due to a recombination cascade. The apparent intensity inversion, which is, to our knowledge, the first observation for a high-density hot plasma, could be due to the opacity effect and/or to charge transfer between B^{5+} ions and neutral atoms.

Resonance charge-transfer recombination [17,18] can take place in the following fashion: the initial flashover takes place along the capillary wall which produces a cylindrical BN-plasma when the ablated material is ionized as the discharge current rises. The cylindrical plasma then implodes toward the capillary axis. At the maximum pinch, the electron temperature (100∼150 eV) rises rapidly, mainly due to shock heating, and it produces a large number of fully stripped boron ions. At the last stage of the implosion, the internal inductance becomes maximum. This induces a high voltage across the capillary electrodes, and the discharge seeks a less inductive current path, which results in an another flashover along the wall. Such a second flashover at maximum pinch has been commonly observed [19] in plasma pinch devices in the past. The plasma, which is pinched at the axis, now expands radially at a velocity close to 5×10^6 cm/sec (ion acoustic velocity) and the streaming B^{5+} ions now interact with the dense neutral atoms which are produced by the second flashover at the wall. Thus, the n=4 and the n=5 states of the B^{4+} ion will be populated rapidly due to resonance charge-transfer recombination, *e.g.*,

$$B + B^{5+} \rightarrow B^{+} + B^{4+} + \Delta Ei \quad (3)$$

where ΔEi is the energy defect between the initial and the final states at infinite separation. Sharp resonances with large peak cross-sections are predicted at low relative velocities, with the energy defect being in the range of 10 to 20 eV [18,20,21]. When the B atom is in the ground state, charge-transfer recombination takes place preferentially to the n=5 and 4 states of the B^{4+} ion for relative velocities of $\sim 10^6$ cm/sec and 10^6 to 10^7 cm/sec, respectively, with cross sections of the order of 10^{-15} cm^2 [20]. Even when the boron atom is in an excited state, charge-transfer recombination also takes place mostly to the n=4 and 5 states [17].

The cross section of quasi-resonant charge-transfer recombination is about 2.5×10^{-15} cm^2 for a Z=5 projectile ion, according to Ref. 21. With an energy defect in the range of 10 to 20 eV, the velocity for the maximum cross-section lies in the range of 10^6 to 10^7 cm/sec. For example, in the charge-transfer recombination with a neutral B from the B^{5+} ion to the n=4 state of the B^{4+} ion, the boron atom, either in the ground state ($1s^2 2s^2 2p$) or in an excited state, satisfies the condition of quasi-resonant charge-transfer recombination because the energy defect is between 10 and 20 eV. This implies that when a boron atom exists, whether it is in the ground state or in an excited state, it experiences quasi-resonant charge-transfer recombination with a B^{5+} ion. This is almost true for charge-transfer recombination with a nitrogen atom (as well as with Si and Ca atoms).

The pumping rate (P_{ct}) for the charge-transfer recombination process with a neutral boron and nitrogen can be estimated by the equation,

$$P_{ct} = N_0 \sigma v \ , \quad (4)$$

where σ is the cross section of the charge-transfer recombination, N_0 is the density of neutral atoms (boron and nitrogen), and v is the relative velocity between B^{5+} and neutral atoms. The second flashover of the discharge produces a dense neutral boron and nitrogen atoms, and the combined neutral atomic density is assumed to be as dense as 10^{19} cm^{-3}. By considering the acoustic plasma velocity (about 5×10^6 cm/sec) of B^{5+} ions in our case, the pumping rate can be roughly estimated to be about 10^{11} sec^{-1}. This value is higher than the radiative decay rate from the n=4 states to the ground state and competes with the rate of collisional mixing for $n \geq 3$ states, which would explain the population density inversion observed in this experiment. In a similar way, it is also expected that charge-transfer recombination occurs preferentially to the n=5 state, but is not likely to occur to the n=3 and 2 states because the charge-transfer recombination to the n=3 and 2 states requires a relative velocity of the order of 10^8 cm/sec.

The high opacity due to the self-absorption between the energy levels in B^{5+} ions could also explain the intensity anomaly among the L_γ, L_δ, L_ϵ, and L_β lines, but not that for the L_α line. A possible assumption to explain the anomaly for all these lines is that during the discharge, a cool plasma plume with a considerable population in the ground state of the B^{4+} ion is likely to extend out from the orifice of the capillary. The plasma is still within the field of view of the spectrograph and contributes mostly to the intensity of the L_α line. Another possible minor cause of the decrease of the L_β intensity would be due to the absorption by carbon deposits on the optical surfaces. However, the difference in the mass absorption coefficient of carbon at 4.1 nm (B V L_β) and at 3.9 nm (B V L_γ) is so small that the absorption by a reasonably thick (a few hundred nm) carbon deposit can not explain the population inversion between the n=3 and n=4 levels.

Figure 5 shows a time-integrated spectrum for a 14-mm BN capillary discharge plasma in the spectral range of 13 to 27 nm, including the B V H_α line. The spectrum was obtained with a single discharge with an input energy density of 2.5 kJ/cm^3. The width of the entrance slit in this case was 22 μm. In this spectral range, the impurity lines from O^{5+}, F^{6+}, Ca^{9+}, and Ca^{11+} ionic species appear to be strong. The two spectral lines at 14.1 and 14.7 nm, designated by Ca XII $2s2p^6 \rightarrow 2s^2 2p^5$ [22–24], are unexpectedly strong. The presence of Ca^{10+} (see Fig. 2) and Ca^{11+} ionic species suggests that the capillary discharge is an efficient device for producing high-temperature plasmas which may possibly be used for investigating collisionally excited Ne-like Ca XI X-ray lasing [25,26].

The intensity of the B V H_α line tends to show a non-linear increase as the capillary length increases. However, the intensity of the B V H_α line is weak compared to that of the B V L_α line and those of the other spectral lines. This feature contrasts with that of a polyethylene capillary discharge [5] where the plasma parameters are optimum for recombination, causing the C VI H_α line to be the strongest line in the spectrum, indicating a high gain-length product. The marked difference in the H_α line intensity between boron and carbon capillary discharges could be largely due to their atomic number difference. The pumping rate of three-body recombination to the upper lasing state scales as Z^8 [27], assuming reasonable plasma parameters for lasing. Also, the limiting electron density for radiation trapping of the lower lasing level scales as $Z^{4.5}$ [27]. The recombination pumping rate to the n=3 state in the B^{4+} ion is then about 4.3 times less than that in the C^{5+} ion, and the limiting electron density for the boron plasma is 2.3 times lower than that in the carbon case. These effects along with the depopulation of the n=3 state by collisional mixing could make the intensity of the B V H_α line much weaker compared to that of the C VI H_α line.

IV. CONCLUSIONS

Boron-nitride capillary-discharge plasmas with an input energy density of $\sim$2 kJ/cm^3 were studied using time-integrated spectral data in the wavelength range of 2.5 to 27 nm. The electron temperature and the electron density were estimated from the boron spectra. The electron temperature measured from the B VI recombination continuum beyond the Lyman series of B^{4+} and from the high-n transitions of the spectral series lines was about 30 eV. However, the presence of a relatively strong Ca XII ion line indicates that the peak electron temperature could be as high as 150 eV. The electron density measured from the Stark width of the B V L_δ line was about 4×10^{19}/cm^3. These plasma parameters suggest that B V H_α amplification based on three-body recombination pumping is not likely. The intensity anomaly observed in this experiment could be due to either an opacity effect or a population inversion caused by resonant charge-transfer between the B^{5+} ions and the neutral atoms.

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