Excited-level populations in C IV in a recombining Θ-pinch plasma

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Absolute intensities of various transitions from $n=3$, 4, 5, 6, and 7 levels in C IV ions have been measured in the recombination phase of a Θ-pinch plasma. The discharge vessel was filled with 1.87 Pa (14 mTorr) of acetylene and 0.07 Pa (0.5 mTorr) of hydrogen gas to generate the plasma. The deduced upper-level populations showed inversions for all levels of $n=4$ and above. An experiment to produce laser gain at 253.0 nm showed only a factor of 6 enhancement in the line intensity, i.e., there was no lasing. The lack of lasing was attributed to the small gain coefficient, to losses in the cavity, and to the reduced transmission of light at the vacuum-interface quartz windows (due to a coating from the plasma).

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The lithiumlike isoelectronic sequence has been found in theoretical studies [1,2] and experimental observations to exhibit population inversions and lasing under suitable conditions of plasma recombination [3–7]. A comprehensive review can be found in Ref. [8]. It has been predicted by theoretical modeling [9] that the plasma conditions in the recombining phase of the Θ pinch at the National Institute of Standards and Technology (NIST—formerly NBS) would be suitable for producing population inversions in C IV. Experiments reported in a previous publication [9] showed population inversions of the $4f$ and $5g$ levels of C IV with corresponding transitions at 116.9 and 253.0 nm in the recombining phase of the plasma. In this Brief Report, the experiments that optimized the production of C IV ions and the measurement of excited-level populations in $n=4$, 5, 6, and 7 are presented. All these excited levels showed population inversions. However, the resultant gain-length products were too small to produce lasing. An attempt to produce lasing at 253.0 nm is also discussed.

The schematic of the experimental setup has been shown in Fig. 2 of Ref. [9]. The details of the Θ pinch and its operation have been described elsewhere [10]. Two different experiments have been conducted; one for absolute excited-state population densities using the spectral-emission measurements, and the other to produce laser gain at 253.0 nm. Three spectrometers, a 1-m normal-incidence (NI) spectrometer, a 2.2-m grazing-incidence (GI) spectrometer, and a ($\frac{2}{3}$)-m air-path Czerny-Turner (CT) spectrometer were used in the first setup for the spectral observations and measurements. The NI was calibrated using an argon miniarc as an irradiance standard [11]. The basic optical setup has been described in Ref. [9]. However, the CT instrument was aligned to observe radiation in two different optical setups: (i) as shown in Fig. 2 of Ref. [9] for plasma ion-temperature measurements, and (ii) a 45° mirror was positioned near the optical axis of the GI spectrometer so that nearly the same plasma volume was viewed by both GT and CT for *in situ* calibration of the GI using the branching-ratio technique [12].

Initial experiments to observe line emission in the recombination phase of the plasma were carried out by filling the discharge vessel with 1.47 Pa (11 mtorr) of a gaseous mixture of hydrogen and 0.05 Pa (0.4 mtorr) of methane (CH_4). The time histories of the C V $1s^2\,{}^1S-1s4p\,{}^1P$ transition at 3.34 nm, observed by using the GI, the C IV $4f$-$3d$ transition at 116.9 nm, observed by using the NI, and the discharge current in the coil, monitored by a Rogowsky loop are shown in Fig. 1. The temporal histories reported in this paper were obtained

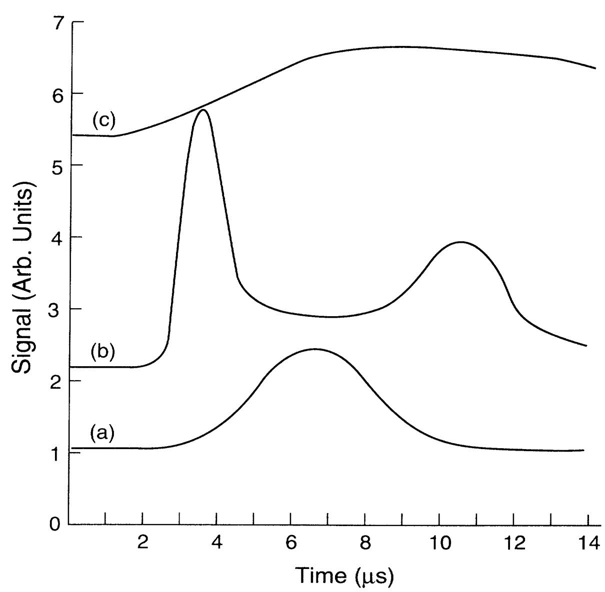

FIG. 1. Time histories of (a) the emission of C V $1s^2\,{}^1S-1s4p\,{}^1P$ transition at 3.34 nm; (b) the emission of C IV $3d$-$4f$ transition at 116.9 nm; (c) the discharge current for a single discharge of the Θ-pinch device.

using a 100-MHz analog-to-digital converter with a 50-ns integration time. Arbitrary units are chosen in Fig. 1, so that the temporal history of the line emission can be compared in relation to the Θ-pinch current. All observed emission lines of each ionic state showed the same characteristic temporal history as in Fig. 1. There is no distortion of these temporal histories from the radiative lifetimes of the upper state of the emission lines since these lifetimes are much shorter than the plasma lifetime for each ionic state in the Θ pinch. Therefore, the temporal histories of emission lines reflect the temporal evolution of ionic populations. The time lag seen between the first peak of the C IV emission line and the C V emission line is due to the ionization time of C IV into C V. The second peak observed in the C IV line emission is due to the recombination of C V ions into C IV [9]. The recombination plasma emission was reproducible in its temporal features within a few percent in the first half cycle of repeated discharges. However, the basic temporal features were also observed in the succeeding two or three half cycles.

The theoretical simulations of Ref. [9] showed that the C IV ionic production in recombination can be maximized by generating plasma conditions of electron density and temperature that produce C V in its ground state, but not energetic enough to excite to higher states or to ionize further the ion. The ionization potential and the first excited state of C V is almost a factor of 6 greater than the ionization potential of C IV (64.5 eV). Consequently, it should be possible to generate C V ions as the dominant ionic species. In order to achieve these conditions, the discharge vessel was filled with a mixture of 18.7 Pa (14 mtorr) of acetylene (C_2H_2) instead of methane and 0.07 Pa (0.5 mtorr) of hydrogen. This increased the carbon fraction in the plasma substantially, as seen by the increase in the C IV line emission. In addition, the discharge voltage of the capacitor bank was lowered (to 15 kV) to reduce further heating of the plasma that would otherwise lead to C V excitation and ionization. The optimum bank voltage was empirically determined by monitoring the decrease in the intensity of the C V resonance line at 4.03 nm. This optimized plasma condition produced the recombination peak (second peak) in C IV early in time with respect to the ionization peak (first peak) as shown in Fig. 2; also, the C V line emission at 4.03 nm became weaker.

Figure 3 shows the temporal evolution of the emission of the observed transitions to the $2p$ level from $3d$, $4d$, $5d$, and $6d$ levels in the optimized plasma condition. The second peak is more intense relative to the first peak for lines originating from higher levels, as expected from recombination, thus giving evidence for the second peak to be due to recombination. It should be noted that the intensity of the first peak would be much larger for $3d$-$2p$ transition if it is corrected for the optical depth. Therefore, the drop in population density of lower levels in recombination is more significant than shown in Fig. 3. No direct measurement of the opacity was made. However, the effect of trapping is not significant for the recombination peaks because of the decrease in density of lower-level populations in the recombination phase.

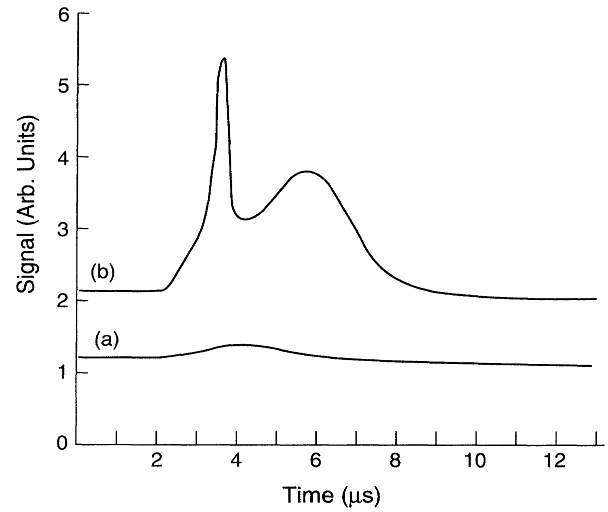

FIG. 2. Time histories of C V and C IV line emission for optimized plasma conditions. (a) $1s^2\,{}^1s-1s2p\,{}^1P$ transition at 4.03 nm; (b) $3d$-$4f$ transition at 116.9 nm.

The emission signal of the recombination peaks was measured for the various transitions shown in Fig. 4. These transitions had measurable signals over the background and were selected because there was no line blending. The absolute intensities (in $\mathrm{W\,cm^{-2}\,sr^{-1}}$) were deduced from these measurements and the absolute calibration of the NI and GI spectrometers. The populations of the excited levels N_i ($\mathrm{cm^{-3}}$) at the peak of the recombination were deduced from the measured absolute emission intensities I_{ik} and the known transition probabilities [13] A_{ik} by using the expression given below for optically thin lines:

$$N_i=(4\pi/h\nu)(I_{ik}/lA_{ik})\ , \qquad (1)$$

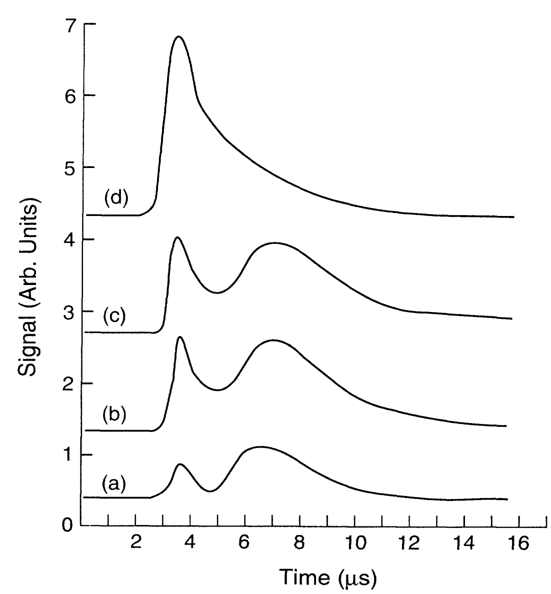

FIG. 3. Time histories of C IV transitions (a) $2p$-$6d$, (b) $2p$-$5d$, (c) $2p$-$4d$, and (d) $2p$-$3d$.

where $h\nu$ is the transition energy, i is the upper level and k is the lower level of the transition, and l is the plasma length in cm. The plasma length has been measured to be 23 cm [10]. The measured population densities of the upper levels are listed in Table I. The upper-level populations deduced from the absolute intensities of branching-ratio lines from the same upper level shown in Fig. 4, measured by using the two different monochromators (NI and GI), were well within the calibration uncertainty of 25%. Therefore, the overall uncertainty in the population densities in Table I was estimated to be $\pm 25\%$. Population inversions between many levels can be seen in Table I. The inversion factor [8] F is given by

$$F = 1 - q_u N_l / g_l N_u \ , \tag{2}$$

where g_u and g_l are statistical weights and N_u and N_l are population densities of upper and lower levels. The maximum measured values of F for $5g$,$4f$ and $4f$,$3d$ levels are 0.6 and 0.9, respectively. A value of F greater than 0.5 showed the possibility of building laser oscillation [8] using a cavity even though the absolute population densities are quite low. However, the ion temperature deduced from the Doppler broadening of the C IV spectral lines was 1 keV, at the optimum plasma conditions [9].

The gain coefficient for a Doppler broadened line can

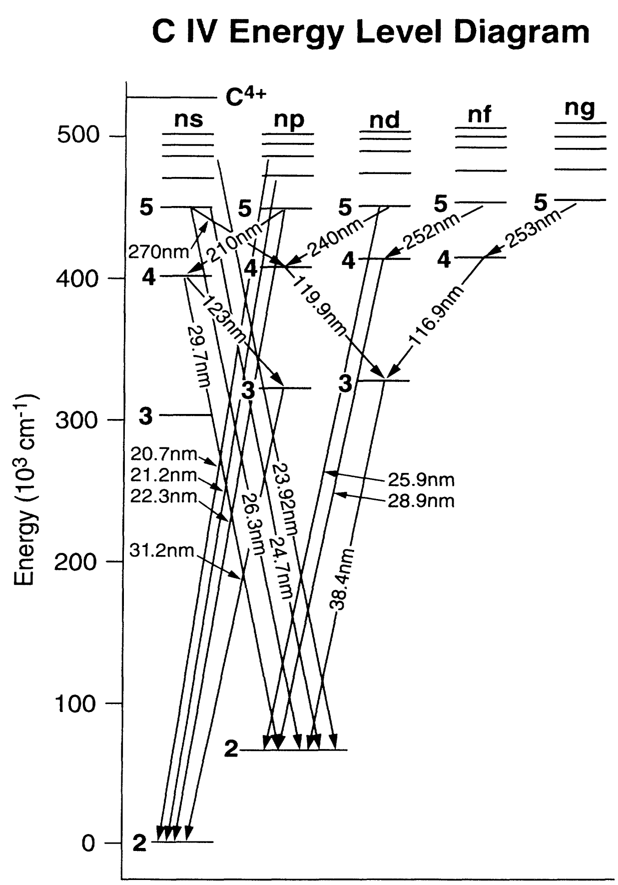

FIG. 4. Grotrian diagram showing transitions measured at the peak of the recombination phase of the plasma.

TABLE I. Upper-level populations at the recombination peak.

Level	Population (10^{10} cm^{-3})
$3p$	1.3
$3d$	2.0
$4s$	6.9
$4d$	5.1
$4f$	13.0
$5s$	5.5
$5p$	14.7
$5d$	8.6
$5f$	9.0
$5g$	28.0
$6s$	12.0
$6p$	18.0
$7s$	20.0
$7p$	13.0

be calculated by using the relation [8]

$$G = 1.7 \times 10^{-8} \lambda^3 A (M/T)^{1/2} N_u F \ , \tag{3}$$

where λ is the wavelength (cm), A is the transition probability (s^{-1}), M is the mass (amu), T is the ion temperature (eV), and F is the inversion factor. The wavelengths and transition probabilities are found in Ref. [13]. The value of G at 243.0 and 116.9 nm for the $5g$-$4f$ and $4f$-$3d$ transitions was 0.008 cm^{-1} and 0.003 cm^{-1}, respectively, at the observed density of level populations due to the high ion temperature. These values of G were only a factor of 3 higher than previously reported [12], even though the plasma has been optimized for maximum recombination line intensities. These low values of G imply the need for a reflective cavity to demonstrate lasing. However, this is presently impractical at 116.9 nm because of the lack of highly reflecting mirrors at this wavelength.

However, the value of $Gl = 0.18$ for $l = 23$ cm at 253.0 nm suggests that a laser could be generated by building a cavity because high reflectivity mirrors are available. An experiment was conducted with mirrors of reflectivity greater than 99% in the rear and about 99% in the front at 253.0 nm to check for the possibility of laser oscillation. Because the line emission during the recombination phase lasted for more than 1 μs, it would make many passes in the cavity. In order to verify this experimentally, the end monochromators were disconnected from the discharge tube and UV transmitting vacuum interface quartz windows were installed at the Brewster angle. Confocal mirrors (1-m focal length) were installed on opposite ends of the discharge tube approximately 1 m apart with adjustable mounts to optimize the optical alignment along the axis of the plasma discharge. A ($\frac{1}{4}$)-m monochromator was installed 2 m away from the partially transmitting (1%) front mirror, and appropriate apertures were used to confine the field of view to the plasma along the axis of the discharge vessel. The linearity of the photomultiplier response was checked with appropriate neutral-density filters. The temporal history of the line intensity at 253.0 nm was observed by a UV-sensitive photomultiplier tube and signals were recorded for plasma discharges, with (I) and without (I_0) the back cavity mirror in the optical path. An enhancement

(I/I_0) by a factor of 5 to 6 was observed. This factor was always a maximum for the first shot after installing clean windows. The windows showed an observable coating in fewer than five shots. Figure 5 shows the time histories for the maximum enhancement observed. The ionization peak showed only a maximum factor of 1.5 increase, which can be accounted for by the reflected intensity from the front mirror. The enhancement during the recombination peak (as well as that at the ionization peak) decreased slowly as more shots were taken. The coating on the windows was a carbon deposit and could not be avoided because of the use of acetylene to generate the plasma. The observed enhancement was degraded because of losses in the cavity and the absorption at the windows. Therefore, we conclude that lasing in this recombining Θ-pinch plasma is not possible because of the large ion temperature, modest populations in the excited levels, cavity losses, and window absorption.

The simulations [12] that prompted these experiments were performed in the collisional radiative model with three-body recombination as the important process. However, previously reported experiments [14] with lithiumlike oxygen and nitrogen in the NIST Θ pinch indicated charge exchange with neutral hydrogen as a possible recombination mechanism. It remains to be determined which process produced the population inversions in C IV observed in the present experiments. An additional series of experiments are planned with the NIST Θ pinch to determine the nature of the recombining mechanism in the acetylene, hydrogen plasma. These may shed some light on the dominant recombination mechanism and will be discussed in a separate paper.

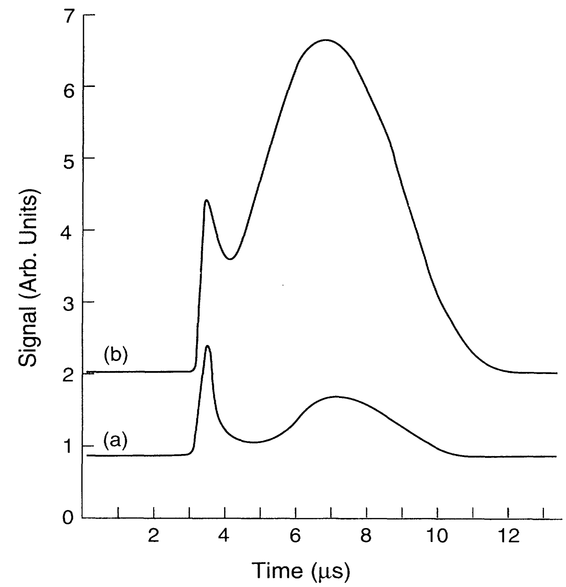

FIG. 5. Time histories of C IV 4f-5g transition (a) without the back mirror and (b) with the back mirror.

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