Electron-impact excitation and recombination into excited states of lithiumlike ions

R. U. Datla and H. -J. Kunze*

Atomic and Plasma Radiation Division, National Bureau of Standards, Gaithersburg, Maryland 20899

(Received 9 October 1987)

Relative excitation rate coefficients of N V and O VI are measured using a well-diagnosed Θ-pinch plasma. Levels of $n=4$ and 5 are included in the measurements for the first time. Experimental values are in satisfactory agreement with theoretical values. A method to deduce the recombination rate coefficient from the He-like ions is suggested from the observations of the effect of recombination on transitions originating from levels of principal quantum number $n=4$ and 5. Recombination rates of $\sim 10^{-12}$ cm^3 s^{-1} at 69 eV for N V and $\sim 10^{-13}$ cm^3 s^{-1} at 40 eV for O VI are deduced. These rates are larger than the known theoretical rates for radiative, three-body, and dielectronic recombination; charge exchange with hydrogen is suggested as possible explanation.

I. INTRODUCTION

The analysis of line radiation emitted by ions in well-diagnosed transient plasmas provides a suitable means to obtain effective electron-ion rate coefficients for ionization, recombination, and excitation. The principles of this experimental method and early results have been discussed previously.[1] A recent review by Griem[2] analyzes all measurements carried out so far. Collisional excitation rate coefficients for ions of the lithium isoelectronic sequence have been reported in Refs. 3–6. In the present investigation we extended such measurements to levels of higher principal quantum number and studied the influence of recombination on the time history of certain transitions which allows the derivation of recombination rates. This is of specific interest to x-ray and uv laser schemes utilizing the recombination approach.

II. THEORETICAL CONSIDERATIONS

At low electron densities, where recombination is negligible, excited states are essentially populated by electron collisions from the ground state (2s for Li-like ions). With increasing electron density the 2p level becomes populated and excitation from this level contributes to excited-state population as well. In general, the emission coefficient ε of an optically thin spectral line $(i \rightarrow f)$ may be written as

$$\varepsilon(i\rightarrow f)=\frac{h\nu}{4\pi}A(i\rightarrow f)N(i)\ , \tag{1}$$

which becomes, for this case,

$$\varepsilon(i\rightarrow f)=\frac{h\nu}{4\pi}\frac{A(i\rightarrow f)}{\sum_{j(<i)}A(i\rightarrow j)}N_e[N(2s)X(2s\rightarrow i)+N(2p)X(2p\rightarrow i)]\ , \tag{2}$$

where $h\nu$ is the photon energy, the A's are transition probabilities, N_e is the electron density, $N(2s)$ and $N(2p)$ are population densities of the respective levels, and the X's are collisional rate coefficients. In this model collisional mixing even between levels of equal principal quantum number and cascading is neglected.

If we assume that all ions in the Li-like stage are essentially in the 2s and 2p levels,

$$N(2s)+N(2p)\simeq N_{\rm Li}\ , \tag{3}$$

and introduce the ratio

$$N(2p)/N(2s)=\beta\ , \tag{4}$$

the emission coefficient in this coronal limit can be written

$$\varepsilon(i\rightarrow f)=\frac{h\nu}{4\pi}\frac{A(i\rightarrow f)}{\sum_{j(<i)}A(i\rightarrow j)}N_eN_{\rm Li}X_{\rm eff}(i)\ , \tag{5}$$

with the effective excitation rate coefficient

$$X_{\rm eff}(i)=\frac{1}{1+\beta}X(2s\rightarrow i)+\frac{\beta}{1+\beta}X(2p\rightarrow i)\ . \tag{6}$$

The ratio β has to be determined experimentally or theoretically and approaches the Boltzmann factor with increasing collisional coupling.

Equation (5) is the central equation of the method and elucidates the necessary experimental measurements. In order to derive an absolute rate coefficient from the emission coefficient, the line intensities must be determined and the concentration of the Li-like ionization stage has to be known. In the case of relative rate coefficients, the product $N_eN_{\rm Li}$ cancels in the ratio requiring only a relative radiometric calibration of the spectrometers. We selected $X(2s\rightarrow 3s)$ as a reference since excitation from 2p to 3s is very small and self-absorption of the emission line $3s\rightarrow 2p$ is negligible.

With increasing principal quantum numbers the levels are also increasingly collisionally coupled to the ground state of the next (He-like) ionization stage, and population by recombination cascade has to be taken into account. This will show up in the temporal evolution of emission lines even during the ionizing phase of the discharge when the species has been essentially ionized through to the next stage. In steady state, the population density $N(x)$ of a level x influenced by recombination from the He-like ion stage is given by

$$N_e X_{\rm eff}(x) N_{\rm Li} + N_e \alpha(x) N_{\rm He} = N(x) \sum_{j(<x)} A(x \to j) , \quad (7)$$

where $\alpha(x)$ is the effective recombination rate coefficient into the specific level x and $N_{\rm He}$ is the density of the ions in the He-like ionization stage. We introduce the ratio ρ of the concentrations in the two successive ionization stages

$$\rho = N_{\rm He}/N_{\rm Li} , \quad (8)$$

and write the emission coefficient from Eqs. (2) and (7):

$$\varepsilon(x \to f) = \frac{h\nu}{4\pi} \frac{A(x \to f)}{\sum_{j(<x)} A(x \to j)} N_e N_{\rm Li} X_{\rm eff}(x) \times \left[1 + \rho \frac{\alpha(x)}{X_{\rm eff}(x)} \right] . \quad (9)$$

The temporal evolution of the emission line will not show the recombination effect as long as

$$\rho \frac{\alpha(x)}{X_{\rm eff}(x)} \ll 1 . \quad (10)$$

On the other hand, in order to determine the influence of the recombination on the population of high-lying levels, we simply compare the time history of an emission line originating at a high-lying level with that from a low-lying level which is not significantly influenced by recombination, i.e., for which Eq. (10) still holds. The intensity ratio R of the two transitions can be written as

$$R = R_0(T) \left[1 + \rho \frac{\alpha(x)}{X_{\rm eff}(x)} \right] , \quad (11)$$

where R_0 is the ratio of line emission coefficients at temperatures where recombination is not observable and T is the electron temperature. The relative change of R with time thus allows the observation of the influence of the recombination on the population of high-lying levels with respect to collisional excitation. The ratio ρ is obtained from computer modeling of the concentration of successive ionic stages occurring in our transient plasma as a function of time. It consists of calculating the line intensities that best fit the observed ones from successive stages of ionization by solving the coupled rate equations governing the transient ionization in our plasma. The main data input for the modeling are the measured electron temperature and density as a function of time. This procedure has been described in detail in literature, especially in Refs. 1–8.

III. EXPERIMENT

The experiment is conducted on the National Bureau of Standards (NBS) Θ-pinch plasma. The details of the operation of this device have been described elsewhere.[7,8] A 2.2-m grazing-incidence monochromator (2.2-m GI) with a holographic grating of 1800 lines/mm is aligned to view the plasma end-on along the axis of the Θ-pinch coil.

It is equipped with a clear plastic scintillator in vacuum and a photomultiplier tube to detect the radiation at its exit slit. The response of this instrument is calibrated *in situ* by using the branching-ratio technique[9] in order to measure the absolute intensities of lines emitted from the plasma ($\rm W\,cm^{-2}\,sr^{-1}$). Line pairs from Ne VIII at 8.8 and 282 nm, O VI at 15 and 381 nm, N V at 20.9 and 462 nm, and C IV at 31.2 and 580.2 nm are used for calibration.

The discharge tube is filled with 22-mTorr of hydrogen gas. It is mixed with 0.5% of either nitrogen or oxygen to study the spectral line emission from N V or O VI ions, respectively. The spectral line intensities as a function of time from $n=3$, 4, and 5 levels for N V and O VI ions are recorded using the 2.2-m GI monochromator. The monochromator viewed the entire 1.5-cm-diameter cross section of the plasma. The measured intensities are line-of-sight averages along the plasma column and are reproducible from shot to shot within 3%. The occurrence of the peak emission in time is reproducible within 10%, whereas the rise and decay times are reproducible on the average to within 30%.

IV. RESULTS AND ANALYSIS

Both temporal and spatial electron temperature and density profiles of the plasma are determined by 90° Thomson scattering of ruby laser light. The spatial profiles are averaged over the cross section of the 1.5-cm diameter of the plasma, and the average electron density and temperature as a function of time are shown in Figs. 1(a) and 1(b), respectively. A 10% uncertainty in the electron temperature and a 25% uncertainty in the electron density are estimated from shot-to-shot variations in the Thomson scattering signals. An ion temperature of 1 keV is measured from the Doppler widths of O VI lines at 381.1-nm using a $\frac{1}{4}$-m monochromator. Hence it follows that during the lifetime of the ionization stage ($\sim 1\ \mu$s) the ions travel collision free a distance of about 5 times the diameter of the plasma column, and a uniform ion distribution within the plasma column can safely be assumed.

The spectral line temporal evolutions are the same for all transitions during the rapid rise to the peak due to the ionization of the previous (Be-like) ion. However, in the recombining period of the plasma, the temporal evolution of transitions arising from higher lying levels, e.g., $n=4$ and 5, deviated from those of the lower-lying levels. This deviation is attributed to recombination from the He-like ion. Typical temporal evolutions of line intensities from the lower-lying and the higher-lying levels of N V and O VI are shown in Fig. 2. The solid lines represent the temporal evolutions of the $3p$-$2s$ transitions and the dashed curves represent the average deviation in the time history observed for the $5p$-$2s$ transitions in N V and O VI. The observed change in the intensity ratio of $5p$-$2s$ and

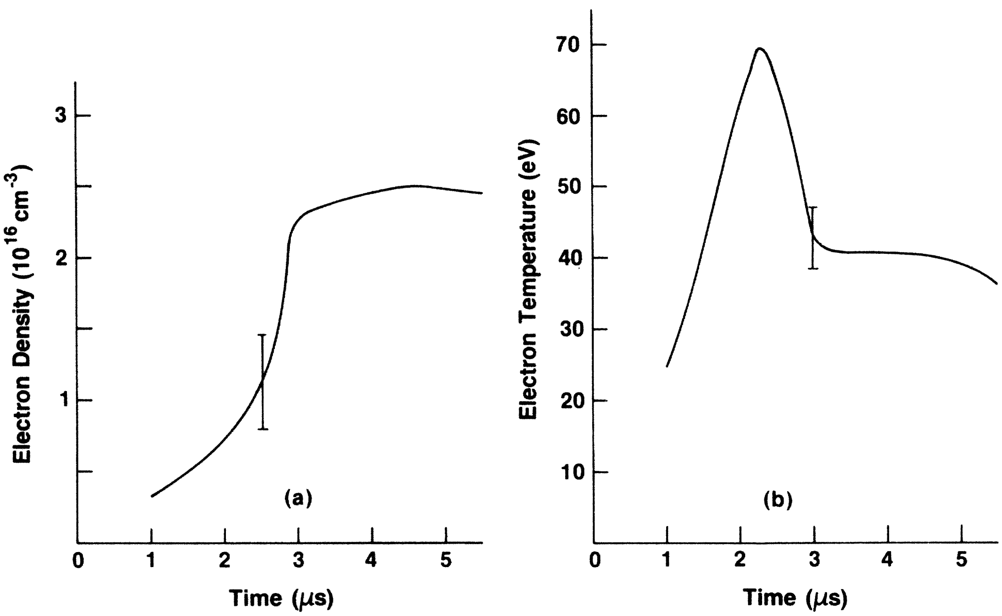

FIG. 1. (a) Radially averaged electron density. (b) Radially averaged electron temperature obtained by ruby laser scattering. The vertical bars indicate the estimated error in each plot.

3s-2p transitions in N V is shown in Fig. 3 as a function of time. The ratio deviates from its (normalized) value of unity starting at 1.9 μs. The deviation increases as the contribution to the intensity of the 5p-2s transition due to excitation from the Li-like ground state reduces because it is depleted by ionization. The bars in Fig. 3 indicate the uncertainty due to shot-to-shot fluctuations.

The relative intensities of various transitions measured absolutely at the peak emission in the temporal evolution are given in Table I. The transition probabilities given in column 3 are taken from available calculations.[10,11] The optical depth corrections to the measured intensities are estimated by using the methods given in Refs. 12 and 13. The correction was 11% and 20% for the 2p-3d transitions and 6% and 7% for the 2s-3p transitions in N V and O VI, respectively. The correction for the rest of the transitions are negligible.

A. Excitation

Table II summarizes the experimentally determined relative excitation rate coefficients. They are deduced from Eq. (5) using the relative intensities given in Table I. The ratio β in Eq. (4) is theoretically evaluated for our plasma conditions by using the semiempirical excitation rate coefficients for $2s \rightarrow 2p$, as given in Ref. 14. It is 0.88 for N V and 0.54 for O VI at the time of peak line intensity where the electron temperatures were 45 and 50 eV, re-

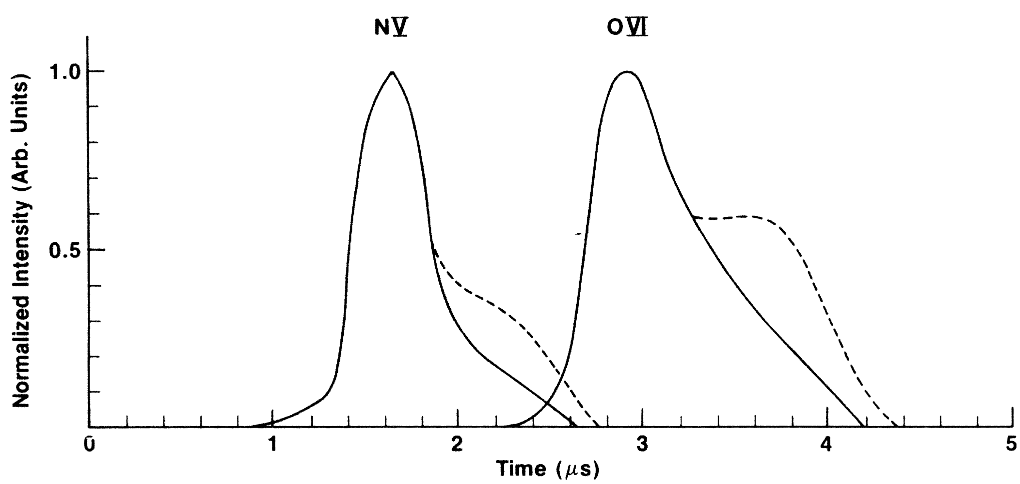

FIG. 2. Intensities of emission lines from $n=3$ levels of N V and O VI normalized to their peak values as a function of time from the initiation of the main bank. The dashed curves represent the 5p-2s temporal evolution at late times in N V and O VI.

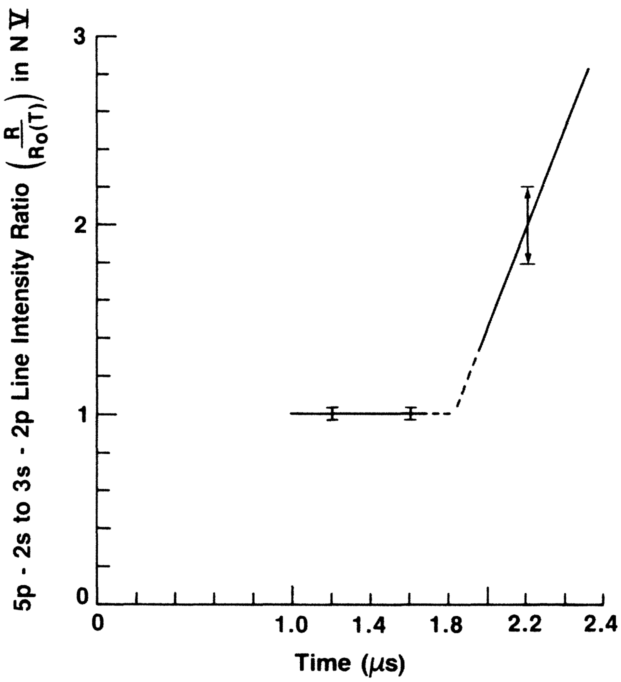

FIG. 3. Intensity ratio $[R/R_0(T)]$ of normalized temporal evolution of 5*p*-2*s* and 3*s*-2*p* transitions in N V.

spectively. The corresponding electron densities were 5.5×10^{15} cm^{-3} and 2×10^{16} cm^{-3}, respectively. The theoretical relative excitation rate coefficients are also given in Table II for comparison. The values given under the column heading of McWhirter are obtained from the semiempirical formulas given in Ref. 14. References 15 and 16 are used in conjunction with Ref. 17 to calculate the rate coefficients listed under the heading Sampson. For N V the rate coefficients are also calculated by using a Coulomb-Born approximation and are listed under the heading Bely.[18,19] The value of the effective theoretical excitation rate coefficient to the 3*s* level in each ion is given in brackets under each heading. There is general agreement among the theoretical results, except in the excitation to 4*p* in N V. The experimental results quoted in the Table II are deduced neglecting collisional coupling within the levels of a given principal quantum number. We have estimated the effect of collisional coupling at our plasma conditions by using the formula given by Griem[2] for excitation transfer within the levels of the same principal quantum number (n) and solving the rate equations in the full collisional radiative model. The calculated population ratios of levels with $n=3$ and 4 in O VI showed that the population of p levels increased while that of s and d levels decreased compared to the calculations based on the pure corona model. The difference was about 25%. The experimental population ratios (Table I) for O VI are closer to that of the collisional radiative model. The effect of collisional coupling was only few percent in the case of N V. This is due to the low electron density of the plasma at the time of the occurrence of the N V ionization stage.

The total uncertainty in the quoted experimental ratios for N V is estimated to be less than ±30%. However, in the case of O VI and for the levels of large n, the error could be ±50%. Our results are in agreement with previous measurements for these transitions reported in the literature.[3,5]

TABLE I. Experimental results and relevant atomic data.

Ion and transition	Wavelength (Å)	Transition probability (10^8 s^{-1})	Relative intensity[a]	Relative upper-level populations[c]
N V				
$5p\rightarrow2s$	104.8	69.4	0.08	0.11
$4p\rightarrow2s$	115.8	126.9	0.43	0.37
$5d\rightarrow2p$	116.4	137.6	0.17	0.13
$4d\rightarrow2p$	129.8	287.0	1.03	0.43
$4s\rightarrow2p$	132.3	64.0	0.10	0.18
$3p\rightarrow2s$	150.1	259.0	3.70[b]	1.78
$3d\rightarrow2p$	173.0	884.0	5.33[b]	1.15
$3s\rightarrow2p$	184.1	170.0	1	1
O VI				
$5p\rightarrow2s$	147.4	33.0	0.07	0.11
$4p\rightarrow2s$	162.6	56.3	0.29	0.28
$5d\rightarrow2p$	166.9	67.4	0.15	0.13
$4d\rightarrow2p$	186.1	140.0	0.63	0.29
$4s\rightarrow2p$	190.2	45.7	0.18	0.25
$3p\rightarrow2s$	209.3	120.0	1.67[b]	1.06
$3d\rightarrow2p$	247.6	429.0	3.50[b]	0.78
$3s\rightarrow2p$	266.2	90.9	1	1

[a]Measurements are with respect to the $3s\rightarrow2p$ transition.
[b]Corrected for optical depth.
[c]These values are independent of the lower state of each transition.

TABLE II. Relative excitation rate coefficients.

Ion, electron temperature T_e, and excited level (i)	Relative X_{expt}	Relative X_{theory} McWhirter	Sampson	Bely
N V at $T_e=45$ eV				
$X_{eff}(5d)$	0.14		0.17	
$X_{eff}(5p)$	0.06		0.07	
$X_{eff}(4d)$	0.58		0.54	0.6
$X_{eff}(4p)$	0.22		0.42	0.25
$X_{eff}(4s)$	0.13		0.12	0.13
$X_{eff}(3d)$	3.7	4.33	3.76	4.8
$X_{eff}(3p)$	1.4	1.44	1.24	1.27
$X_{eff}(3s)$	1	1	1	1
		$(5.49\times10^{-10})^a$	$(5.41\times10^{-10})^a$	$(5.63\times10^{-10})^a$
O VI at $T_e=50$ eV				
$X_{eff}(5d)$	0.15		0.19	
$X_{eff}(5p)$	0.06		0.08	
$X_{eff}(4d)$	0.96		0.64	
$X_{eff}(4p)$	0.34		0.19	
$X_{eff}(4s)$	0.08		0.14	
$X_{eff}(3d)$	5.02	5.8	5.05	
$X_{eff}(3p)$	2.75	1.70	1.67	
$X_{eff}(3s)$	1	1	1	
		$(2.77\times10^{-10})^a$	$(2.75\times10^{-10})^a$	

[a]These are absolute excitation rate coefficients from theory. The units are $cm^3\,s^{-1}$.

B. Recombination

The ratio ρ in Eq. (9) is obtained by computer modeling of the ionic concentrations using the experimental plasma electron temperature and electron density as input.[3] Figure 4 shows the relative concentrations of N V and N VI thus calculated. The ratio α/x_{eff} in Eq. (9) is deduced from the measured value of $R/R_0(T)$. The recombination rate coefficient α is then estimated by using the effective excitation rate coefficient given by the calculations of Sampson.[16] The results are shown in Table III. The values are very sensitive to the accuracy of ρ. At late times, ρ can get very large as there is very little population of Li-like ions. So it is best to choose a time where ρ is not larger than 80–85 and $R/R_0(t)$ is not much less than about 2. It is difficult to estimate the error in the experimental values of recombination rate coefficients thus obtained because ρ is deduced by theoretical (computer) modeling, and there are fluctuations in the signals especially when the intensities are small. The uncertainty could be as high as a factor of 2.

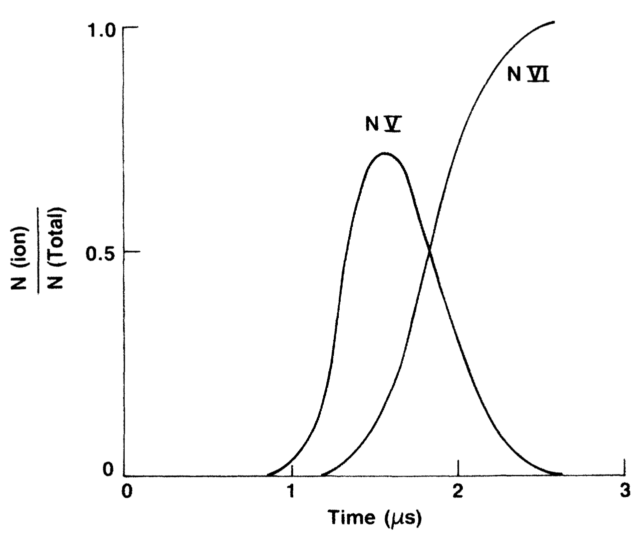

FIG. 4. Calculated temporal evolution of the fractional abundance of N V and N VI. N (ion) refers to the density of N V or N VI. N (Total) refers to the total ion density.

The interpretation of the results, however, poses some problems since the recombination rates are at least factor of 10 larger than predictions based on three-body[13] or dielectronic recombination formulas.[20] Furthermore, the ionization rate coefficients from the ground state of N V and O VI are at least a factor of 100 larger than these recombination rates, which explains why substantial recombination to the lithiumlike ground state is not observed.

We propose, therefore, that charge exchange recombination of the heliumlike ion is responsible. It would occur preferentially into levels of specific quantum numbers. The effective rate coefficient is written[21]

$$\alpha^{cx}=\frac{N_H}{N_{H^+}}\langle v\sigma_{cx}\rangle , \qquad (12)$$

where σ_{cx} is the respective cross section. The cross sections given in Refs. 22 and 23 require a density ratio of neutral hydrogen atoms to protons of the order of 10^{-5} to explain the observed recombination rates. Theoretical estimates[24] yield a ratio of $N_H/N_{H^+}\simeq10^{-7}$. These estimates do not take into account the possibility of the ions

TABLE III. Recombination rate coefficient measurements.

Ion observed transition	Temperature (eV)	ρ	$R/R_0(T)$	X_{eff} ($cm^3 s^{-1}$) Sampson	α_{expt} ($cm^3 s^{-1}$)
N V					
$5d \rightarrow 2p$	69	34	2.52	9.98×10^{-11}	4.5×10^{-12}
$5p \rightarrow 2s$	69	34	2.52	4.03×10^{-11}	1.8×10^{-12}
$4p \rightarrow 2s$	69	34	1.4	2.49×10^{-10}	2.9×10^{-12}
$4s \rightarrow 2p$	69	34	2.5	$5.4\ \times10^{-11}$	2.3×10^{-12}
O VI					
$5d \rightarrow 2p$	40	10.5	1.4	1.78×10^{-11}	6.8×10^{-13}
$5p \rightarrow 2s$	40	10.5	1.4	$7.3\ \times10^{-12}$	2.8×10^{-13}
$5s \rightarrow 2p$	40	10.5	1.6	$3.1\ \times10^{-12}$	1.8×10^{-13}

interacting with the cool gas surrounding the plasma. It is conceivable in our plasma case that the ions, being hot (~ 1 keV), can charge exchange with the neutrals at the plasma boundary. Neutrals that have not been ionized and swept up by the pinch effect, and the neutrals released from the walls, could form into a cool blanket around the plasma. No estimate of this density is available but it could be sufficiently large to account for the ratio of $N_H/N_{H^+} \simeq 10^{-5}$.

V. SUMMARY

Relative excitation rate coefficients for transitions up to levels of principal quantum number $n=5$ are measured for Li-like N V and O VI ions. The measurements for $n=4$ and 5 levels are done for the first time. They are compared with theoretical calculations and satisfactory agreement is found. For O VI, collisional coupling between different levels of the same n affected the measured ratios by 25%. The recombination rate coefficients are estimated from the observed deviations of the temporal evolutions of spectral lines arising from $n=4$ and 5 levels when compared to $n=3$ levels.

ACKNOWLEDGMENTS

The authors thank Dr. J. R. Roberts and Dr. D. H. Oza for helpful discussions.

*Permanent address: Ruhr-Universität, 4630 Bochum, West Germany.

[1]H.-J. Kunze, Space Sci. Rev. **13**, 565 (1972).
[2]H. R. Griem, J. Quant. Spectrosc. Radiat. Transfer (to be published).
[3]H.-J. Kunze and W. D. Johnston III, Phys. Rev. A **3**, 1384 (1971).
[4]H.-J. Kunze, Phys. Rev. A **4**, 111 (1971).
[5]G. N. Haddad and R. W. P. McWhirter, J. Phys. B **6**, 715 (1973).
[6]C. C. Chang, P. Greve, K.-H. Kolk, and H.-J. Kunze, Phys. Scr. **29**, 132 (1984).
[7]W. L. Rowan and J. R. Roberts, Phys. Rev. A **19**, 90 (1979).
[8]R. U. Datla and J. R. Roberts, Phys. Rev. A **28**, 2201 (1983).
[9]E. Hinnov and F. W. Hoffman, J. Opt. Soc. Am. **53**, 1259 (1962).
[10]W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities,* Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 4 (U.S GPO, Washington, D.C., 1966), Vol. 1.
[11]G. A. Martin and W. L. Wiese, J. Phys. Chem. Ref. Data **5**, 537 (1976).
[12]R. C. Elton and W. W. Köppendörfer, Phys. Rev. **160**, 194 (1967).
[13]H. R. Griem, *Plasma Spectroscopy* (McGraw-Hill, New York, 1964).
[14]D. M. Cochrane and R. W. P. McWhirter, Phys. Scr. **28**, 25 (1983).
[15]L. B. Golden, R. E. H. Clark, S. J. Goett, and D. A. Sampson, Astrophys. J. Suppl. Ser. **45**, 603 (1981).
[16]R. E. H. Clark, D. H. Sampson, and S. J. Goett, Astrophys. J. Suppl. Ser. **49**, 545 (1982).
[17]D. H. Sampson provided the σ values for use in $Z_{eff}=Z-\sigma$ for various transitions in Li-like ions needed for the calculations (private communication).
[18]O. Bely, Proc. Phys. Soc. London **88**, 587 (1966); Ann. Astrophys. **29**, 131 (1966); **29**, 683 (1966).
[19]O. Bely and D. Petrini, Astron. Astrophys. **6**, 318 (1970).
[20]S. M. Younger, J. Quant. Spectrosc. Radiat. Transfer **29**, 67 (1983).
[21]H. C. Meng, P. Greve, H.-J. Kunze, and T. Schmidt, Phys. Rev. A **31**, 3276 (1985).
[22]R. A. Phaneuf, F. W. Meyer, and R. H. McKnight, Phys. Rev. A **17**, 534 (1978).
[23]D. H. Crandall, R. H. Phaneuf, and F. W. Meyer, Phys. Rev. A **19**, 504 (1979).
[24]H. R. Griem, in *Physics of Plasmas Close to Thermonuclear Conditions,* edited by B. Coppi, G. G. Leotta, D. Pfirsch, R. Pozzoli, and E. Sidorii (Commission of the European Communities, Brussels, 1980), Vol. I.