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Photoionization spectrum of the ^3P ground state of neutral carbon

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The atomic absorption of C I has been studied in the 1100–385-Å spectral region, using the flash-pyrolysis method to produce the absorbing layer and a BRV as a continuum source. Thirty-four lines belonging to the $2s^22p^2\,^3P - 2s2p^2np\,^3D^o$ series converging to the $2s2p^2\,^4P$ ionization limit have been observed and for the first time resolved in fine structure. In addition, energies of $np\,^3D^o_{1,2,3}$ are deduced from these observed lines. Three new lines have been observed and identified as simultaneous excitation of the two outer electrons to the final state $2s^2\,3pnd\,^3D^o$. The relative photoionization cross section of the ground state ^3P, as measured in the same range of wavelengths, is found to be consistent with theoretical predictions.

I. INTRODUCTION

Carbon has repeatedly been the subject of experimental spectroscopic interest since 1923.[1] Up to now the most complete analysis has been made by Johansson.[2] He summarized all the previous works and considerably extended the knowledge of C I spectrum by a list of 450 observed lines and 301 computed wavelengths, ranging from the infrared down to the $2s^22p\,^2P^o$ ionization limits.

Through data recorded during a solar flare by the Naval Research Laboratories (NRL) spectrograph on Skylab, the list of predicted wavelengths has been almost completely confirmed and further extended.[3] Esteva *et al.*[4] used an original absorption technique to observe a few autoionized lines in the photoionization continuum. Mainly because of experimental difficulties these are the only observations of this kind carried out so far. In fact, the C I atomic-emission spectra with low-excitation light source, are strongly blended with molecular bands, while a higher-excitation energy dissociates the molecules but produces ionized carbon. On the other hand, in absorption the traditional laboratory techniques are not able to dissociate the carbon compounds.

This paper discusses absorption spectra of C I obtained by the method of flash pyrolysis, which turned out to be the right tool in order to dissociate and vaporize the sample and at the same time to produce clean atomic spectra. The examined region is between the first ionization limit at about 1100 and 385 Å, the lower limit of efficiency of our grating. The relative analysis allows us to identify 34 lines, practically all new. They are $2s^22p^2\,^3P–2s2p^2np\,^3D^o$ series converging to the $2s2p^2\,^4P$ ionization limits. For the first time the fine structure has been resolved. Furthermore three new features have been observed and identified as double excitation transitions from the ground state to $2s^23pnd\,^3D^o$ levels converging to $2s^23p\,^2P^o$ limits. The photoionization cross section is measured in the same wide region 1100–385 Å, supporting experimentally what was formerly known only by theoretical means.

II. EXPERIMENT

The absorption spectra of C I have been obtained with the pyrolysis technique in the spectroscopy laboratory of the Arcetri Observatory employing the instrumentation previously used in the experiments concerning the spectra of litium,[5] silver,[6] and gold.[7] Spectra are recorded by a two-meter normal incidence spectrograph equipped with a holographic grating whose dispersion is 4.17 Å mm^{-1}.

The absorbing vapor is produced with the flash-pyrolysis system consisting of a xenon flash lamp powered by low-inductance 120-μF capacitor operated up to 9 kV. As a sample we had 99.99% pure graphite with grains of about 30-μm size. After a delay of about 120 μs we fired the continuum source, a modified BRV[8] powered by a 2.5-μF capacitor charged to 20–22 kV. The continuum radiation was focussed by a toroidal mirror which enabled us to record one spectrum with a single shot on Kodak 101-05 plates or Kodak 101-01 film, using 15-μm-wide slit.

Because of the large cross section of the graphite powder in the vacuum ultraviolet, even a small quantity of such grains remaining in the cloud of carbon atoms absorbed most of the background continuum so that low-intensity spectra and hence poor information on C I resulted. For this reason we have used the pyrolysis system with which it is possible to heat the sample to about 5000 °C, a temperature slightly higher than that needed for graphite dissociation. Good results have been achieved with a preheating of the graphite powder by means of a low-powered flash shot. In this way gases trapped in the grains are released and a fragmentation of the grains themselves is produced. For measurement of the photoionization cross section we cut off the second-order radiation of the grating by means of rare gases acting as filters. Moreover Ar, Kr, and Ne absorption lines have been used as standards to calculate the wavelengths of carbon lines.

A reasonable number of spectra were obtained. From these the best two or three using each kind of gas were analyzed in order to increase the reliability of the weak lines and to give internal consistency on all results. For well defined lines we have a precision of the order of 0.01 Å. Spurious absorption and emission lines, mainly of neutral and ionized carbon, oxygen, and uranium, originating in the continuum source, sometimes reduced the accuracy of our measurements.

It is evident from our data that for the absorption spectrum of carbon the pyrolysis technique is superior to previously used ones. While we observed no substantial difference between the pyrolysis and induction furnace spectra in our studies of elements like Ag and Au (Refs. 6 and 7), it is clear that traditional methods (King furnace, etc.) cannot supply the high temperature needed for the evaporation of carbon.

III. ANALYSIS OF THE SPECTRA

A. Resonances

The electronic configuration of C I is $1s^2 2s^2 2p^2$. According to Moore (Ref. 9), $^3P_1$, $^3P_2$, $^1D_2$, $^1S_0$ lie, respectively, 16.42, 43.42, 10 192.66, and 21 648.02 cm^{-1} above the ground $^3P_0$ level. The absorption of photons with a wavelength shorter than 1100 Å by neutral carbon atoms in the ground state might give rise to bound-free transitions of one electron or to autoionizing transitions of one or more electrons.

These transitions are well studied from a theoretical point of view because of their great interest in astrophysics and in the atomic structure. A recent theoretical work by Carter and Kelly,[10] predicted the trend of the continuum photoionization cross section with superimposed autoionized lines originating in transitions from the ground levels ^3P to $2s2p^2(^4P, ^2P)np\,^3D^o$, $^3P^o$, $^3S^o$, $2s^2 3s3p\,^3P^o$, and $2s^2 3p3d\,^3D^o$, $^3P^o$. They computed also, for all these lines, the resonance energy and the Fano parameters Γ and q, predicting also the series $2s2p^2(^4P, ^2P)np\,^3S^o$ to be very weak with respect to the others and consequently very difficult to be observed experimentally. Other important theoretical works are those of Taylor and Burke[11] and Burke and Taylor,[12] where a different computational approach is used. They evaluated energies of $2s2p^2(^4P)np$ resonances and calculated continuum cross sections which are in good agreement with values given by Carter and Kelly,[10] while discrepancies there are in other values of Fano parameters. Burke and Taylor[12] also computed the energies of limits for the ion in the $2s2p^2\,^2D$, ^2P, ^2S, and $2p^3\,^4S^o$, $^2D^o$, $^2P^o$ configurations. Rydberg series converging to such limits are expected to be very weak and therefore hardly observable.

The first experimental results in this region came from works of Edlén[13] and Boyce and Rieke.[14] They all observed the three emission lines corresponding to transitions from the fine structure of the ground state to $2s2p^3\,^3S_1^o$ level. Esteva *et al.*,[4] using a vacuum spark to generate the absorption medium, observed and measured lines due to transitions from the ground state to $2s2p^2(^4P)np\,^3D^o$, with n ranging from 3 to 7 and to $np\,^3S^o$ with $n = 2, 3$. Nevertheless, for both the poor homogeneity of absorbing vapor and for the low-resolving power of the spectrograph, no fine structure has been detected.

In our experiment we observed 37 lines, which are shown arranged in series in Tables I and II. These tables give, for each measured line, the principal quantum number n, the intensity I, which is a visual estimate on a scale from one to nine, the wavelength λ, the wave number ν, and the effective principal quantum number n^*.

It should be noted that the value of λ represents the wavelength of the absorption peak, which is not coincident with the resonance wavelength in the case of an autoionized line with asymmetrical profile. In our observed lines we have a negative profile index q implying that λ (*resonance*) $<\lambda$ (*measured*). This may explain the systematic rise with n of the quantum defect $\delta = n - n^*$, resulting especially for the most extended series. The bulk of lines observed are identified as transitions from the ground state $2s^2 2p^2\,^3P_{0,1,2}$ to $2s2p^2(^4P)np\,^3D^o_{1,2,3}$ and are shown in Table I(a)–I(f). The values of the ^4P limits are derived from excited C II levels.[9] According to the selection rules we may expect six lines in each multiplet. In our spectra we may resolve all six lines only in the transition with $n=4$, corresponding to the maximum of separation among $^3D^o_{1,2,3}$ levels. For lines with $n=3$ we observe a strong broadening due to autoionization completely masking any fine structure.

The multiplet corresponding to $n=5$ is shown in Fig. 1, where it is possible to distinguish only five lines and to see on the lower wavelengths side, the typical asymmetrical profile with the "window" of minimum cross section emerging over the continuum. From Fig. 1 it is easy to understand that, because of the proximity of multiplet lines, it is impossible to measure the true profile of lines to deduce Fano's parameter Γ and q and the exact λ of the resonance.

TABLE I. Absorption series to $1s^2 2s2p^2\,{}^4P$ limits.

(a) $2s^2 2p^2\,{}^3P_0$–$2s2p^2np\,{}^3D_1^o$. Limit ${}^4P_{1/2}$ 133 823.72 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1560.3095[a]	64 089.8	1.254
3	(?)[b]	869.3	115 035.1	2.417
4	3	803.15	124 509.7	3.432
5	5	780.13	128 183.8	4.411

(b) $2s^2 2p^2\,{}^3P_1$–$2s2p^2np\,{}^3D_1^o$. Limit ${}^4P_{1/2}$ 133 823.72 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1560.7088[a]	64 073.4	1.254
3	(?)[b]	869.3	115 035.1	2.418
4	3	803.24	124 495.8	3.433
5	5	780.24	128 165.7	4.410
6	3	768.93	130 050.8	5.405
7	3	762.52	131 144.1	6.419
8	3	758.64	131 814.8	7.421
9	2	756.17	132 245.4	8.382
10	1	754.39	132 557.4	9.370
11	1	753.08	132 788.0	10.376
12	1	752.13	132 955.7	11.352
13	1	751.42	133 081.4	12.295

(c) $2s^2 2p^2\,{}^3P_1$–$2s2p^2np\,{}^3D_2^o$. Limit ${}^4P_{3/2}$ 133 845.72 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1560.6820[a]	64 074.6	1.254
3	(?)[b]	869.3	115 035.1	2.416
4	5	802.81	124 562.5	3.441

(d) $2s^2 2p^2\,{}^3P_2$–$2s2p^2np\,{}^3D_1^o$. Limit ${}^4P_{1/2}$ 133 823.72 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1561.3668[a]	64 046.4	1.254
3	(?)[b]	869.3	115 035.1	2.419
4	3	803.37	124 475.6	3.434
5	6	780.40	128 139.4	4.411
6	4	769.08	130 025.5	5.406
7	3	762.69	131 114.9	6.416
8	2	758.76	131 794.0	7.433
9	1	756.31	132 220.9	8.388

(e) $2s^2 2p^2\,{}^3P_2$–$2s2p^2np\,{}^3D_2^o$. Limit ${}^4P_{3/2}$ 133 845.72 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1561.3400[a]	64 047.5	1.254
3	(?)[b]	869.3	115 035.1	2.418
4	7	803.00	124 533.0	3.441
5	5	779.93	128 216.6	4.432

(f) $2s^2 2p^2\,{}^3P_2$–$2s2p^2np\,{}^3D_3^o$. Limit ${}^4P_{5/2}$ 133 874.02 cm^{-1}

n	I	λ(Å)	ν(cm^{-1})	n^*
2		1561.4382[a]	64 043.5	1.254
3	(?)[b]	869.3	115 035.1	2.416
4	4	802.71	124 578.0	3.444
5	6	779.68	128 257.7	4.437
6	3	768.57	130 111.8	5.432
7	4	762.35	131 173.3	6.426
8	4	758.51	131 837.4	7.420
9	3	755.96	132 282.1	8.418
10	2	754.21	132 589.1	9.401
11	1	752.93	132 814.5	10.392
12	1	751.99	132 980.5	11.361
13	1	751.24	133 113.3	12.368
14	1	750.68	133 212.6	13.324

[a] From Johansson (Ref. 2).

[b] Very broad and fine-structure blended; accuracy is ±0.2 Å.

Table III reports energies of $2s2p^2({}^4P)np\,{}^3D^o_{1,2,3}$ levels as deduced from observed lines. For the other two possible series $2s^22p^2\,{}^3P$–$2s2p^2({}^4P)np\,{}^3P^o$, ${}^3S^o$ converging to the same limits as the previously considered series, we observe only three very weak lines at about 945 Å, corresponding to transitions to ${}^3S_1^o$ from the three ground-state levels ${}^3P^o_{0,1,2}$. These lines are not reported in Tables I and II because they are already well known.[2,13,14]

TABLE II. Double-electron transitions $2s^22p^2\,{}^3P$–$2s^23pnd\,{}^3D^o$. Limit ${}^2P$ 222 552.2 cm^{-1}.

n	I	λ(Å)	(cm^{-1})	n^*
3	4	485.58	205 939.3	2.570
4	3	468.37	213 506.4	3.483
5	2	460.81	217 009.2	4.449

An impurity absorption line, due to the continuum source, similar to that identified by Esteva *et al.*[4] as the $2s^22p^2\,{}^3P$–$2s2p^2({}^4P)3p\,{}^3S^o$ carbon line occupied the same position on our spectra. Since we have used the continuum source essentially of the same kind as Esteva *et al.*,[4] we think they also observed the impurity line instead of the identified carbon line. For the resonance transitions ${}^3P$–${}^3P^o$ predicted by Carter and Kelly[10] and Burke and Taylor,[12] in positions corresponding to $n = 3, 4$, we find larger absorption features of the source stronger than any possible carbon line. For $n > 4$ no evidence of lines exists, nor is there evidence of any other possible transitions, except for the three lines shown in Table II and, for one of these, also in Fig. 2. These lines are identified as double-electron transitions from the ground state to $2s^23p({}^2P^o)nd\,{}^3D^o$ because they fit in a series converging to the limit $2s^23p\,{}^2P^o$ whose energy is de-

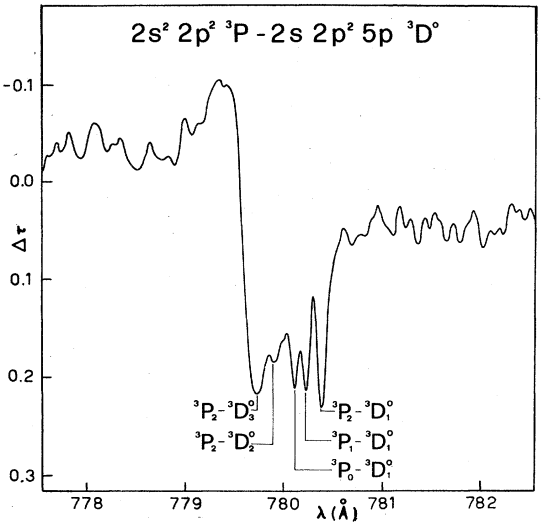

FIG. 1. Profile of optical depth of inner-shell transitions. Absorption increases with $\Delta\tau$. The adjacent continuum corresponds to $\Delta\tau=0$. On the side of shorter wavelengths the window of minimum cross section is easily visible.

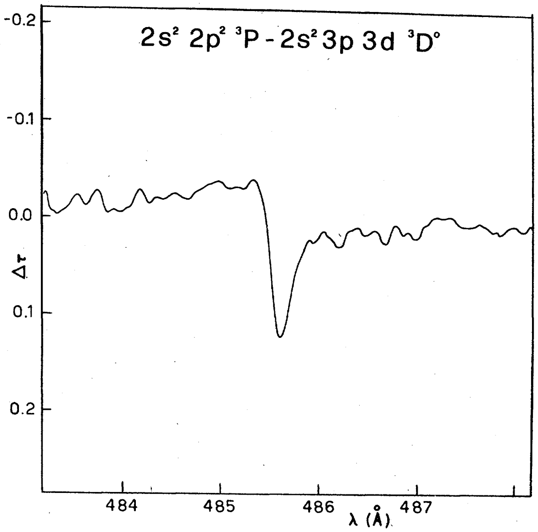

FIG. 2. Profiles of optical depths of double-electron transitions. Absorption increases with $\Delta\tau$ and this has the same scale as Fig. 1. Although the profile is a blend of six lines, the asymmetry of the profile is easily visible.

rived from values of the C II excited level.[9] From an observational standpoint these lines are new. We believe that our line at energy 25.53 eV, having $n=3$, corresponds to the line at 26.35 eV predicted by Carter and Kelly[10] with the same excited core. No fine structures have been resolved for these lines, but we think that the splitting of almost ground ^3P levels surely broadens lines even if there is not any fine-structure evidence. Since each of these three lines is a merging of six lines, no measurement was carried out from Fano's parameters.

B. Photoionization cross section

The relative photoionization cross section has been determined in the wavelength range 1100–385 Å by subtracting from the atomic-carbon continuum-absorption spectrum the background radiation of the BRV source.

TABLE III. Energy levels of $1s^22s2p^2np\,^3D^o$ (cm^{-1}).

	$^3D^o_1$	$^3D^o_2$	$^3D^o_3$
$2p$	64 089.8	64 090.7	64 086.9
$3p$	115 035[a]	115 035[a]	115 035[a]
$4p$	124 513.7	124 577.7	124 621.4
$5p$	128 182.9	128 260.1	128 301.2
$6p$	130 068.1		130 155.2
$7p$	131 159.5		131 216.8
$8p$	131 834.3		131 880.8
$9p$	131 263.1		132 325.6
$10p$	132 573.9		132 632.5
$11p$	132 804.4		132 857.9
$12p$	132 972.2		133 023.9
$13p$	133 097.8		133 156.7
$14p$			133 256.0

[a] Blended.

In case of plane one-dimensional purely absorbing medium the optical depth $\tau(\lambda)$ is defined by

$$\tau(\lambda)=\ln[I_0(\lambda)/I(\lambda)]\,, \tag{1}$$

where $I_0(\lambda)$ and $I(\lambda)$ stand, respectively, for the incident and emerging intensities in the absorption cell at any wavelength λ. Moreover, $\tau(\lambda)$ is related to the atomic photoionization cross section $\sigma(\lambda)$ by

$$\tau(\lambda)=\int_l \sigma(\lambda)N\,dl=\sigma(\lambda)\langle N\rangle_{\rm av}\,l\,, \tag{2}$$

where N is the atomic density and l is the thickness of the medium. Both intensities $I_0(\lambda)$ and $I(\lambda)$ have been measured in relative units, and $I(\lambda)$ has been scaled so that the coefficient $\tau(\lambda)$ is zero at wavelengths longer than the first ionization limit, i.e., for $\lambda>1101.07$ Å, where no photoionization from ^3P may occur. This scaling has been done to take into account the scattering mainly caused by the residual graphite grains which is reasonably independent of λ in this spectral region. To be sure that we were recording just the pure first-order continuum, we have taken several plates with convenient amounts of different noble gases in the absorption cell. In this way the contribution of higher-orders continuum was cut out by Ar at $\lambda\gtrsim786$ Å, by Ne at $\lambda\gtrsim575$ Å, and by He at $\lambda\gtrsim504$ Å. For shorter wavelengths no gas was needed due to the already mentioned very low reflectivity of the grating.

Each spectrum has been scanned with a computer controlled microphotometer, so that digital readings were directly stored in files. The coefficient $\tau(\lambda)$ was derived with high resolution in a semiautomatic way by means of the calibration curve, derived by using several meshes with known transmissions. Special care was taken to account for the different amounts of stray light on each spectrum which may indeed by a problem for measurements in regions with high transparency. In this way we obtained the $\tau(\lambda)$ profile. Since $\tau(\lambda)$ from relation (2) is proportional to $\sigma(\lambda)$, it is possible to derive that value of the proportionality constant $\langle N\rangle_{av} l$ through which it is possible to match the experimental $\tau(\lambda)$ curve and the $\tau(\lambda)$ curve theoretically predicted by Carter and Kelly and Burke and Taylor. The best agreement of the computed $\sigma(\lambda)$ with the ratio $\tau(\lambda)/\langle N\rangle_{av} l$ was found with $\langle N\rangle_{av} l$ value equal to about 10^{16} atoms cm^{-2}. Obvious deviations from one spectrum to the other, were observed with about the same quantity of graphite, but order of magnitudes are maintained. Figure 3 shows the photoionization cross section in absolute units of the ground states ^3P, as well as the resonances and the marked step on threshold $2s2p^{2}\,{}^4P$. In this figure the cross section predicted by means of the R-matrix method[12] is shown as a dot-dashed curve, while theoretical results from the many-body perturbation theory[10] are displayed as the dotted line (length form) and dashed line (velocity form).

There is consistency with an experiment carried out with a wall-stabilized arc by Hofmann and Weissler[15] as regards absolute results in the range 1101–980 Å. In Fig. 3 there are represented samples of these measures (dots); intermediate values lie on the joining segment, with a gap between about 1090–1025 Å, due to spurious lines. Although values are slightly high around the threshold, the agreement is within the estimated experimental errors. A check on the reliability of the $\langle N\rangle_{av} l$ value has been made on the basis of known oscillator strength of the C I multiplet at $\lambda \simeq 1280$ Å. This structure, easily visible in our spectra, arises from the transitions from the ground levels to $2s^2 2p4s\,{}^3P^o$. Its total oscillator strength is

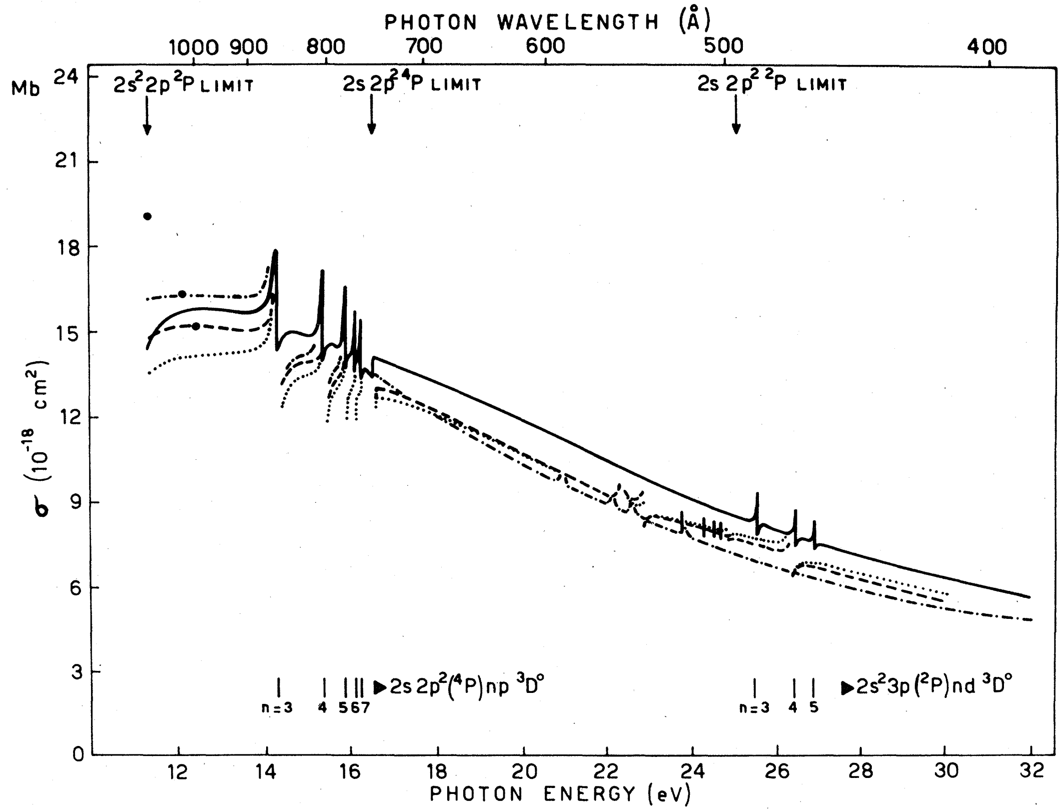

FIG. 3. The measured relative photoionization cross section is shown as a full line in comparison with the theoretical predictions made by the R-matrix method (dot-dashed line), and the many-body perturbation theory [dotted line (length form) and dashed line (velocity form)]. The three dots are from previous measurements using a wall-stabilized arc. The jump of the cross section at the $2s2p^{2}\,{}^4P$ limit is clear together with the converging resonances; only those with n up to seven have been reported. At shorter wavelengths there are three observed resonances from double-electron excitation transitions.

known with an uncertainty of about 10% from measures of Brooks *et al.*[16] The profile of the six lines forming the multiplet can be described by means of six Doppler profiles, because in our conditions ($T \simeq 5500$ °K, $\langle N\rangle_{\mathrm{av}} l \simeq 10^{16}$ atoms cm^{-2}) the damping contribution is negligible. We assumed that *f*-value ratios are the ones according to *LS* coupling rules. The theoretical intensity $I_c(\lambda)$ is given by

$$I_c(\lambda)=I_0(\lambda)\exp\left(-\sum_{j=1}^{6} g_j\sigma_j(\lambda)\langle N\rangle_{\mathrm{av}} l\right), \tag{3}$$

where g_j are the statistical weights of the levels from which transitions start and $\sigma_j(\lambda)$ is the Doppler-broadened cross section for the j line of the multiplet

$$\sigma_j(\lambda)=\frac{\pi e^2}{m_e c^2}\frac{\lambda^2}{\Delta\lambda_D} f_j \exp\left(-\frac{(\lambda-\lambda_{0j})^2}{(\Delta\lambda_D)^2}\right), \tag{4}$$

with λ_{0j} resonance wavelengths of lines, f_j their oscillator strengths, and $\Delta\lambda_D$ the Doppler width, equal about to 0.01 Å, while other symbols have the usual meaning. In order to reproduce the experimental shape of multiplet lines we must convolve the function (3) with the instrumental profile $T(\lambda)$. The latter is evaluated by normalizing the profile of some sharp line appearing in the spectra. The absorption profile, in relative units, is thus

$$\tau_c(\lambda)=\ln\frac{\sum_n I_c(\lambda-\lambda_n)}{I_0}T(\lambda_n), \tag{5}$$

where I_0 is constant in the wavelength interval of the multiplet and it has to be matched with the observed one by adjusting $\langle N\rangle_{\mathrm{av}} l$. The agreement between the values of $\langle N\rangle_{\mathrm{av}} l$, obtained with this and the previously described method, turned out very well. Since $\langle N\rangle_{\mathrm{av}} l \simeq 10^{16}$ atoms cm^{-2} and the geometrical depth is roughly equal to 10 cm, $\langle N\rangle_{\mathrm{av}} \simeq 10^{15}$ atoms cm^{-3}.

Sources of errors are estimated to be (i) the nonperfect reproducibility of the intensity slope versus wavelength of the BRV which is evaluated on a statistical basis, (ii) the impurities present in the absorbing vapor, (iii) the different responses of the photographic emulsions for different wavelengths, and (iv) the neglect of photoionization absorption arising from $^1D_2$. An evaluation of this last effect can be made by deducing the population ratio of $^1D_2$ and ^3P ground levels supposing that collisions are the dominant process, being $\langle N\rangle_{\mathrm{av}} \simeq 10^{15}$ atom cm^{-3}. Therefore levels have Boltzmann population. The fine structure of ^3P is populated according to degeneration because of their small splitting energy with respect to kT. For the $^1D_2$ we have

$$\frac{N(^1D_2)}{N(^3P)}=\frac{g(^1D_2)}{g(^3P)}\exp\left(\frac{-\Delta E}{kT}\right)\simeq 0.038\,. \tag{6}$$

The photoionization cross section for $^1D_2$ at the limit $2s^2 2p\,^2P^o$ is measured from Hofmann and Weissler[15] to be about 9.5 Mb. With the population ratio (6) the contribution to the absorption from $^1D_2$ is 0.4 Mb, equivalent to an error of about 2.5%. The overall uncertainty of the present result on $\sigma(\lambda)$ in Fig. 3 is estimated to be about 30%.

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