$$\frac{\delta E}{\delta \varphi_i'^{\dagger}(x)} = \sum_j \int \frac{\delta E}{\delta \varphi_j^{\dagger}(x')} \cdot \frac{\delta \varphi_j^{\dagger}(x')}{\delta \varphi_i'^{\dagger}(x)}\, dx' = \sum_j \frac{\delta E}{\delta \varphi_j^{\dagger}(x)}\, a_{ji}^{-1*} .$$

By using Eqs. (3.4) and (5.12) one may show that $\hat{\varphi}_i' = \sum_j \hat{\varphi}_j a_{ji}^{-1*}$. Hence, the operator M in the identity $\delta E/\delta\varphi_i^{\dagger} = M\hat{\varphi}_i$ is a scalar and $\delta E'/\delta\varphi_i'^{\dagger} = \delta E/\delta\varphi_i^{\dagger}$ implies $M' = M$. The transformation properties also explain why the derivative with respect to a direct adjoint orbital leads to a scalar operator which operates on a reciprocal rather than a direct orbital.

[47] If a single atomic shell is used for a fragment, the dissociated, or "undistorted" fragment is obtained by shrinking all inner shells into the nucleus and stripping off all outer shells. The reconstruction of the parent system must then be done by switching on the interaction between the shells rather than by bringing the atomic nuclei together.

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PHYSICAL REVIEW A VOLUME 6, NUMBER 2 AUGUST 1972

Energies and Lifetimes of Doubly Excited States in He I

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(Received 28 December 1971)

Using the beam-foil technique we have observed radiative transitions between doubly excited states of He I. From previous calculations, we have been able to classify many of these transitions. Some of the lifetimes of the upper states were obtained from decay-time measurements. There is good agreement between theory and experiment for both wavelengths and radiative lifetimes. A transition between doubly excited states in Li II has also been observed, and we compare experiment and theory for higher members of the He I isoelectronic sequence.

I. INTRODUCTION

The two-electron system provides a good testing ground for many variational and perturbation theories, but until the photoabsorption experiments of Madden and Codling[1,2] in helium, few results were available where both electrons were excited. Such doubly excited states can be reasonably collected into two groups: one group which, under Coulomb selection rules (no change of parity, S, L, or J) can autoionize to an adjacent continuum with the ejection of an Auger electron; and a second group which is stable against autoionization (in the Coulomb approximation) and is of the type $2pnl\ ^{1,3}L$ ($l = L \geq 1$, 1, $n \geq 2$). These states are not situated in continua with the same quantum numbers L, J, S, and parity.

Madden and Codling[1,2] obtained accurate energies for many autoionizing, doubly excited $^1P^{\circ}$ states. Some of these states have also been observed as resonances in electron-helium[3] and ion-helium[4] inelastic scattering. However, the limited energy resolution 0.1–0.5 eV of these latter experiments have limited classifications of the resonances mostly to the $2l2l'$ states. The problem of classification can be seen from considering the number of terms available for the lowest doubly excited configurations. Thus, the $2l2l'$ configuration provides six terms (five autoionizing, one nonautoionizing), and the $2l3l'$ configuration provides 20 terms (16 autoionizing, and four nonautoionizing). These last 20 terms in helium all lie within 1.5 eV of each other.

The nonautoionizing states should be more easily observable by photon emission. However, only transitions from the two lowest states $2p^2\ ^3P$ and $2p3p\ ^1P$ were observed by Kruger[5] and by Compton and Boyce.[6] The classification of the two transitions observed in the extreme vacuum uv were not verified by theory for more than thirty years.[7–9] However, the transition from $2p^{2\,3}P$ has recently been measured very accurately,[10] and its wavelength is in excellent agreement with the theory.[11]

The rapid multiple collisions in the beam-foil excitation technique are efficient at producing multiexcited electronic states,[12] and we have observed a number of other doubly excited states in He I through photon emission in the extreme ultraviolet to singly excited states.[13] In this study, we have made an analysis of the beam-foil spectrum of helium in the uv and visible wavelength regions to search for transitions between nonautoionizing doubly excited states.

II. EXPERIMENT

We accelerated beams of He^+ to energies between 0.25 and 1.0 MeV in a 2-MV Van de Graaff accelerator, and after magnetic deflection directed them

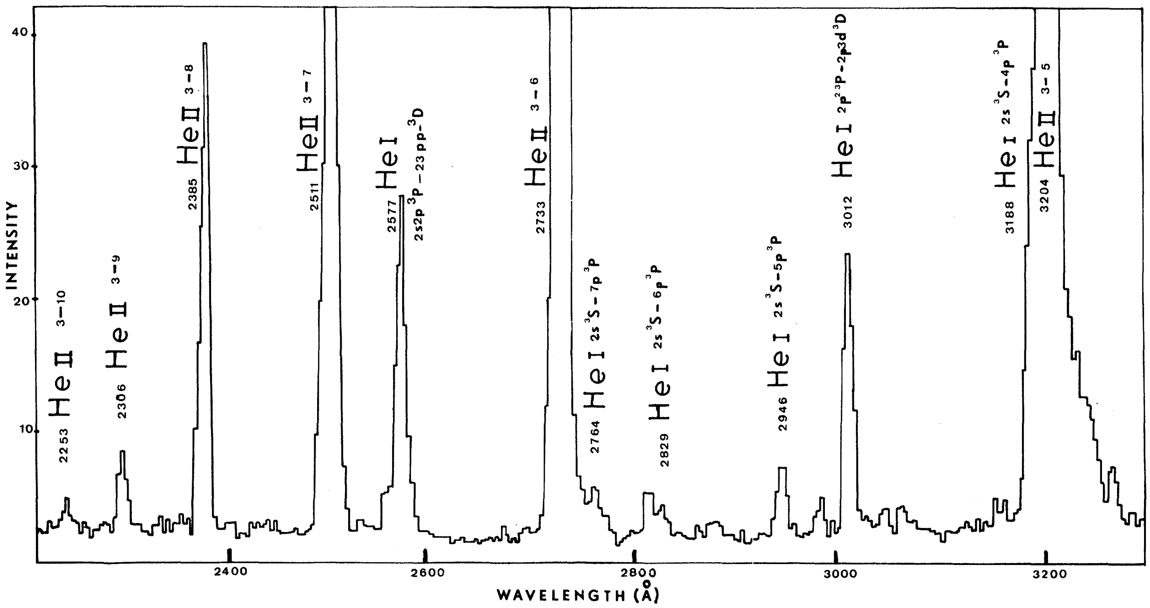

FIG. 1. Partial spectrum of foil-excited helium at 300-keV beam energy. Some identifications are given. A doppler-shifted reflection can be seen at 30-35 Å higher wavelength of each strong line.

through a thin carbon foil (typically 10-μg cm^{-2} thickness). The radiation emitted by the foil-excited atoms and ions was observed at approximately 85° in the forward direction to the beam axis with a 60-cm Czerny-Turner monochromator. Two interchangeable gratings were used, of 1200 lines/mm, and blazed at 2500 and 5000 Å. A double-quartz-lens optical system focused the beam onto the entrance slit with unit magnification; the beam length viewed was thus dependent on the slit width and was typically 0.25 mm. A cooled EMI 6256 photomultiplier was mounted at the exit slit, and the photoelectron pulses were amplified and recorded in a multichannel analyzer. Spectra were observed over the wavelength region 2000–6000 Å. The spectrum between 1100 and 3500 Å was also observed using a 1-m normal-incidence vacuum spectrometer equipped with an EMR F-542 Ascop photomultiplier. The background vacuum was $\sim 4\times 10^{-6}$ torr.

Decay times were measured by moving the carbon foil at constant speed along the beam axis, while its position was synchronized to channel number in the multiscalar. Repeated scans were then made to eliminate effects of variations of beam current and foil conditions during single scans. The scanning speed was 4 mm/min.

III. RESULTS

A. Helium Spectrum

A partial spectrum obtained from a foil-excited helium beam is shown in Fig. 1. The He II spec-

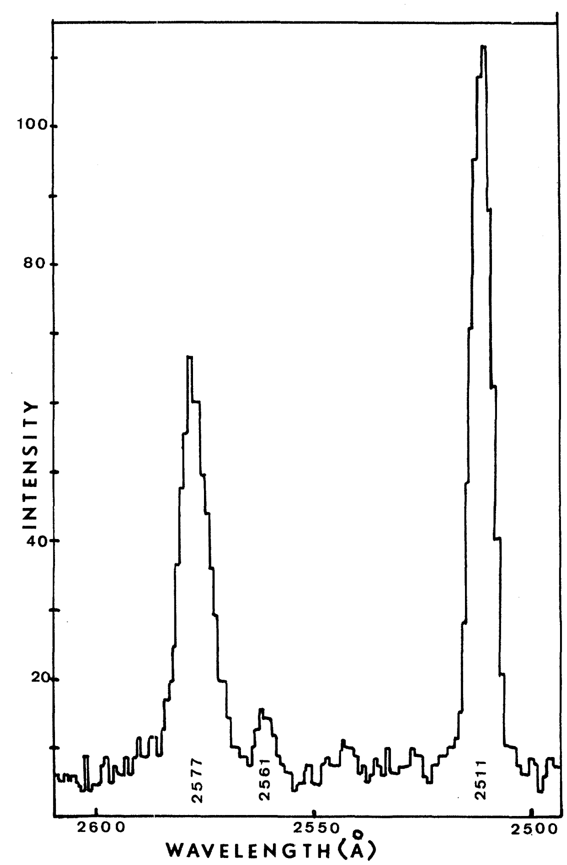

FIG. 2. Linewidth of the He I transition $2s2p\,^3P^o - pp23-{}^3D$ at 2577 Å is compared with the width of the He II transition at 2511 Å. The increased width of the former is due to the rapid autoionization of the lower term.

TABLE I. Observed transitions.

Wavelength (Å) Expt.[a]	Wavelength (Å) Theory	Intensity[b]	Possible classification	Mean life of upper term (nsec) Expt.	Mean life of upper term (nsec) Theory
3470 ± 3	3440[c]	2	$2p^2\,{}^3P$-$sp23-{}^3P^\circ$	...	0.01[d]
3372 ± 2	3492[e]	3	$2p^2\,{}^1D$-$2p3d\,{}^1D^\circ$	...	0.10[f]
3013.7 ± 0.3	3014.0[g]	100	$2p^2\,{}^3P$-$2p3d\,{}^3D^\circ$	0.11 ± 0.02	0.10[f]
2885 ± 1	2990[h]	2	$2p^2\,{}^1D$-$2p3d\,{}^1P^\circ$	...	0.42[d]
2818.2 ± 0.3	2824[c]	20	$2p^2\,{}^3P$-$2p3d\,{}^3P^\circ$	0.15 ± 0.05	0.15[d]
2577.6 ± 0.3	2572	120	$2s2p\,{}^3P^\circ$-$pp23-{}^3D$	0.14 ± 0.02	0.46[d]
2561 ± 1	...	12	$2p^2\,{}^3P$-$2p4d\,{}^3D^\circ$	...	...
2491 ± 2	2494[c]	3	$2p^2\,{}^3P$-$2p4d\,{}^3P^\circ$	...	0.25[d]
2402 ± 1	...	2	$2p^2\,{}^3P$-$2p5d\,{}^3D^\circ$	...	...
2363.9 ± 0.5	2368.8[g]	6	$2s2p\,{}^3P^\circ$-$2p3p\,{}^3P$	0.12 ± 0.04	0.10[e]
2319 ± 1	...	1	$2p^2\,{}^3P$-$2p6d\,{}^3D^\circ$	...	...
2279 ± 3	...	0.5	$2p^2\,{}^3P$-$2p7d\,{}^3D^\circ$	...	...

[a]Wavelength in air.
[b]On a linear scale, not adjusted for detection efficiency.
[c]Lower term Ref. 10, upper term Ref. 15.
[d]Reference 15, autoionization rate only.
[e]Lower term Ref. 17, upper term Ref. 22.
[f]Reference 22, only radiative decay allowed.
[g]Reference 22.
[h]Lower term Ref. 17, upper term Ref. 15.

trum is strong at all energies while the intensities of He I transitions from singly excited states rapidly decrease in intensity with increasing beam energy. These spectra are very well known and have been listed by Martin.[14]

Red-shifted satellites were noted at 25–35 Å higher wavelengths of each strong line. These appeared only when the foil was within the field of view of the monochromator. The satellites are believed to arise from photons emitted from the beam in the backwards direction and then scattered by the foil surface towards the monochromator. The wavelength shifts of the satellites are proportional to the beam velocity and the wavelength of the parent line in accord with this suggestion.

However, other lines were observed and some of these are indicated in the figure. We believe they arise from doubly excited levels in neutral helium. The possibility of ghost or impurity lines was checked by using different beams and exciter foils, e.g., foils of Au, Al, and Ag, whose neutral resonance lines were observed. The C I line at 2478 Å, the C II lines at 4074 and 4269 Å, and molecular bands of N_2^+ at 3914 and 4278 Å and CH at 4300 Å were also seen. A different beam, such as lithium, reproduced the foil spectra, but did not produce the supposed doubly excited helium lines. The two strongest unknown transitions around 3000 Å were also observable with the 1-m normal-incidence spectrometer which has a lower efficiency in this region.

Although the beam of $^4He^+$ was mass-energy analyzed, it also contained a small fraction of oxygen (possibly $^{16}O^{4+}$) which gave rise to the strongest transitions of O II, O III, and O IV excited by the foil. Their intensities were weak and varied relative to the intensities of the helium transitions.

B. Classification

1. General Considerations

We expect to observe radiative transitions from states which have zero or very small autoionization rates. Otherwise, the states will be rapidly depopulated by electron emission. We have included in the term diagram of Fig. 4 the six $2l2l'$ terms and 18 $2l3l'$ terms (the ^1F and ^3F terms have not been calculated and are omitted). The calculated

TABLE II. Alternative classifications of lines at 293.8 and 2577 Å.

Observed wavelength (Å)	Possible classification	Theoretical wavelength (Å)	Mean life (nsec) Upper level Expt.	Upper level Theory	Lower level Expt.	Lower level Theory
293.8	$1s2s\,{}^1S$-$sp23-{}^1P^\circ$	294.0	0.12	4.7×10^{-3} [a]	...	...
	$1s2p\,{}^3P^\circ$-$pp23-{}^3D$	293.9		0.46 [b]	...	...
2577	$2s^2\,{}^1S$-$sp23-{}^1P$	2574	0.14	4.7×10^{-3}	7.0×10^{-5}	4.7×10^{-6} [a]
	$2s2p\,{}^3P^\circ$-$pp23-{}^3D$	2577		0.46	±0.5	6.2×10^{-5} [a]

[a]Reference 15.
[b]Reference 17.

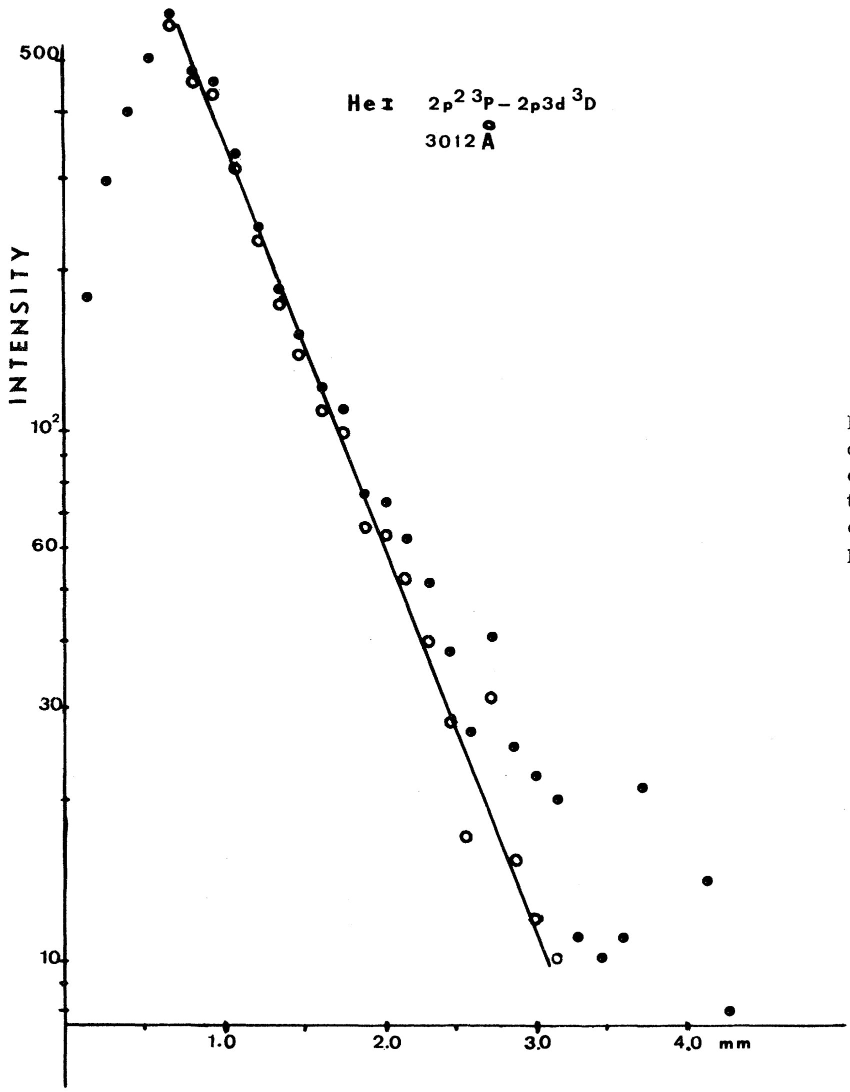

FIG. 3. Decay curve of the He I $2p^2\,{}^3P$-$2p3d\,{}^3D$, 3013 Å transition observed at 300 keV. The closed circles represent the initial data, and the open circles represent the decay curve after subtraction of photomultiplier dark current.

energies are taken from Burke and McVicar,[15] and from Lipsky and Russek[16] for the autoionizing ${}^{1,3}S$ and ${}^3P°$ terms, from Cooper *et al.*[17] and from Altick and Moore[18] for the ${}^{1,3}D$ terms, and from Drake and Dalgarno,[19,20] from Holøien and Midtal,[21] and from Doyle, Oppenheimer, and Drake[22] for the nonautoionizing ${}^{1,3}P$ and ${}^{1,3}D°$ terms. The experimental results of Madden and Codling[2] are used for the ${}^1P°$ terms.

Through configuration interaction, the ${}^{1,3}P°$ terms become linear combinations of $2snp$, $2pns$, and $2pnd$ configurations and they have been extensively investigated both theoretically[15,16,23] and experimentally.[1,3] Most of them can autoionize too rapidly (approximately $10^{-14}S$) for the states to be observable by photon emission. Similar situations occur for the ${}^{1,3}S$ terms which are linear combinations of the $2sns$ and $2pnp$ configurations,[15,16] and for the ${}^{1,3}D$ terms which are linear combinations of the $2pnp$, $2snd$, and $2pnf$ ($n > 4$) configurations.[17,18] However, in all these series, the triplet terms autoionize less rapidly than the singlets, and in some cases the autoionization rates become comparable to the expected radiative decay rates.

From the above considerations, we have been able to suggest classifications for most of the suspected helium lines, as given in Table I.

2. *Linewidths*

As pointed out previously (24), the natural linewidth of a transition connecting one or two autoionizing levels may become sufficiently large to be detectable in beam-foil spectroscopy. In these experiments the smallest instrumental linewidth used is 4 Å and enables us to detect natural linewidths greater than 1 Å. In addition, the instrumental function is closely Gaussian and symmetric in shape, as measured from the profiles of He II lines

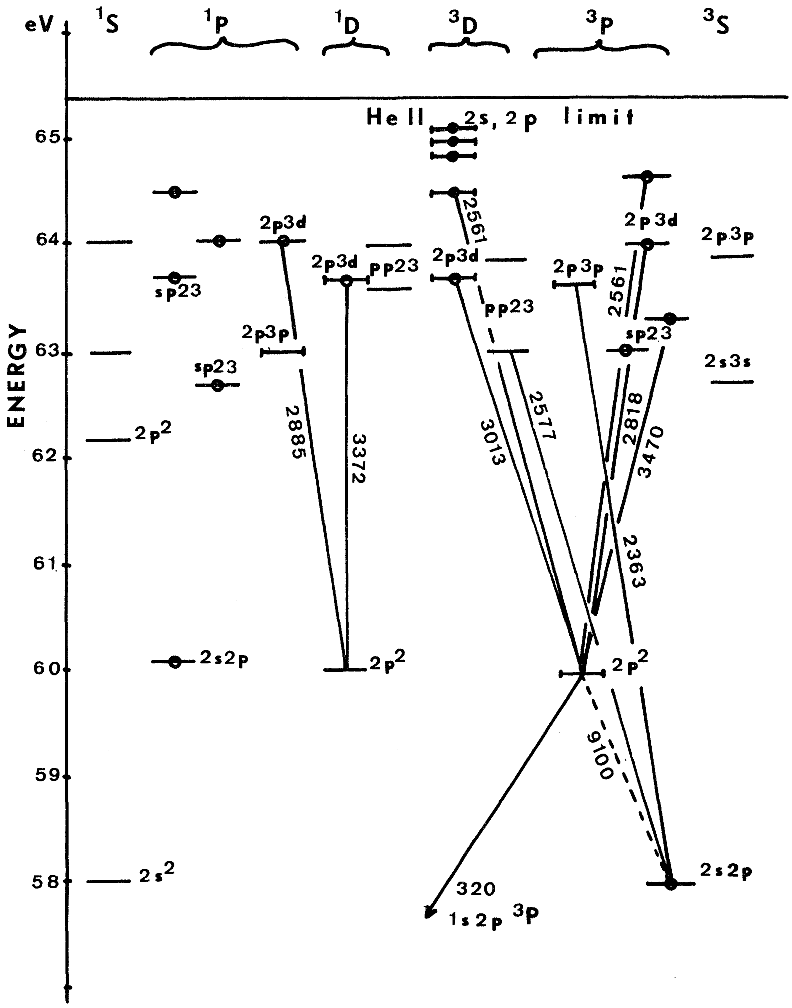

FIG. 4. Energy level diagram of some doubly excited levels below the $2s\,^2S_{1/2}$ ionization limit of He I. The wavelengths of classified transitions observed in beam-foil spectra are indicated in Å.

emitted by the beam; whereas, the strong wings of the Lorentzian natural line shape can be clearly seen, for example in Fig. 2.

The linewidth was measured for each suspected doubly excited helium transition and was deconvoluted assuming a Voigt profile. The resulting natural width, which is the sum of the inverse lifetime of both the upper and lower levels, is compared with theory in Table II.

3. *Discussion of Transitions*

The transition at 3013 Å evidently corresponds to the transition $2p^2\,^3P$-$2p3d\,^3D$ which has just been calculated by Doyle *et al.*[22] to lie at 3014 Å. Its measured lifetime of 0.11 ± 0.02 nsec, shown in Fig. 3, is also in accord with theory.[22]

We have previously suggested a classification[24] for the other strong transition at 2577 Å as $2s^2\,^1S$-$sp23-{}^1P$. This was based partly on our earlier observation in the vacuum uv of the $sp23-{}^1P$ term and on the theoretical wavelength for this transition.[15] However, we show in Table II that the alternative classification of $2s2p\,^3P$-$pp23-{}^3D$ appears more in agreement with theory. The autoionization rate of this upper term is very slow[17,18] and the observed linewidth corresponds very well to the calculated width[15] of the lower term $2s2p\,^3P^\circ$. The origin of the vacuum uv line also becomes more plausible.

The line at 2363 Å had a similar natural width which we attribute to the same lower term, and thus classify the transition as $2s2p\,^3P$-$2p3p\,^3P$ in close agreement with the calculated wavelength.[22,25] The upper term was also observed in the vacuum uv[26] through the transition $1s3p\,^3P^\circ$-$2p3p\,^3P$ at 306 Å. The nonappearance of the transition $1s2p\,^3P^\circ$-$2p3p^3\,P$ at 291.1 Å[21] in those spectra has been explained by Drake[22] by its very low transition probability. The next transition in this series $1s2p\,^3P^\circ$-$2p4p\,^3P$ has been calculated[21] to have a wavelength of 285.0 Å, and this classification appears more probable for the line observed at 285 ±1 Å in Ref. 13.

The transition at 2818 Å had 0±1 Å natural width,

TABLE III. He I Isoelectronic sequence. Comparison of measured and theoretical wavelengths (in Å).

	$1s2p\,{}^1P$-$2p^2\,{}^1D$		$1s2p\,{}^3P$-$2p^2\,{}^3P$		$1s2s\,{}^3S$-$2s2p\,{}^3P$	
	Theory	Expt.	Theory [a]	Expt.	Theory	Expt.
He I	319.8 [b]	...	320.27	320.27 [c]	322.22 [d]	...
Li II	141.2 [b]	...	141.0	...	140.8 [d]	...
Be III	78.87 [b]	78.92 [e]	78.66	78.662 [e]	78.53 [d]	78.550 [e]
B IV	50.17 [b]	50.22 [f]	50.05	50.05 [f]	49.94 [d]	49.945 [f]
C V	34.77 [g]	34.70 [f,h]	34.60	34.586 [f]	34.51 [d]	34.525 [f,h]
N VI	25.46 [g]	...	25.34	...	24.65 [d]	...
O VII	19.43 [g]	19.421 [f,i]	19.35	19.366 [f]	19.30 [d]	... [i]
Ne IX	12.345 [j]	12.355 [j]	12.34	...	12.303 [j]	12.303 [j]
Mg XI	8.548 [k]	8.550 [k]	8.55	...	8.519 [k]	8.519 [k]
Si XIII	6.263 [k]	6.265 [l]	6.27	...	6.244 [l]	...

[a]References 9 and 22.
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[i]Also observed by N. V. Roth and R. C. Elton, NRL Report No. 6638 (unpublished).
[j]Reference 29.
[k]Reference 30.
[l]J. F. Meekins, G. A. Doschek, H. Friedman, T. A. Chubb, and R. W. Kreplin, Solar Phys. 13, 198 (1970).

and there is little doubt in its classification as the $2p^2\,{}^3P$-$2p3d\,{}^3P^\circ$ transition.

The higher members of the series of transitions $2p^2\,{}^3P$-$2pnd\,{}^3D^\circ$ ($n=4$, 5, 6, 7) were found by assuming that the quantum defects of the upper terms were close to 0.10. No calculations have been published for these terms.

Two transitions between doubly excited singlets are observed: The transition $2p^2\,{}^1D$-$2p3d\,{}^1D^\circ$ at 3470 Å and the transition $2p^2\,{}^1D$-$2p3d\,{}^1P^\circ$ at 2879 Å. The upper term of the latter is calculated to autoionize very slowly[15] in contrast to the other ${}^1P^\circ$ terms.

C. Isoelectronic Sequence

Many calculations have been made for doubly excited states of the heavier ions in the He I isoelectronic sequence, and a few experimental results are available. We have observed a line in lithium which is close in wavelength to a transition suggested by Dalgarno.[27] It is the $2s2p\,{}^3P$-$2p^2\,{}^3P$ transition at 5510 ± 3 Å whose predicted wavelength is 5584 Å. The line has a half-width of 20 ± 10 Å in reasonable agreement with the predicted width of 17 Å, due to the autoionization of the lower level. The corresponding transition in He I should be about 9100 Å and has not been observed. In the higher members of the isoelectronic sequence, Edlén and Tyrén[28] first observed transitions from the doubly excited states as faint satellites to the Lyman-α lines of the respective single electron spectra. These transitions have since been seen also in plasma discharges[29] and in observations of the solor corona.[30] In Table III, we have compared the observed spectra with a number of recent calculations for the lowest doubly excited states. The agreement is quite remarkable, although in Ne IX, Mg XI, and Si XIII, there may be some blending present.

Recently, Jalufka and Cooper[31] have raised doubts on the origin of these satellite lines. They have shown that argon impurity lines overlap in wave-

TABLE IV. Term energies.

	Energy (eV)		
Upper term	This expt.	Theory	
$sp23-{}^3P^\circ$	63.244 ± 0.003 [a]		63.276 [b]
$2p3d\,{}^3P^\circ$	64.0703 ± 0.0005 [a]		64.121 [b]
$2p4d\,{}^3P^\circ$	64.647 ± 0.005 [a]		64.645 [b]
$2p3d\,{}^3D^\circ$	63.7849 ± 0.0004 [a]		63.7854 [c]
$2p4d\,{}^3D^\circ$	64.511 ± 0.002 [a]		...
$2p5d\,{}^3D^\circ$	64.831 ± 0.003 [a]		...
$2p6d\,{}^3D^\circ$	65.016 ± 0.003 [a]		...
$2p7d\,{}^3D^\circ$	65.110 ± 0.004 [a]		...
$pp23-{}^3D$	63.116 ± 0.001 [d]	63.141 [e]	63.157 [f]
$2p3p\,{}^3P$	63.551 ± 0.002 [d]		63.555 [g,h]
$2p^2\,{}^1D$	59.988 ± 0.005 [h]	60.025 [e]	60.115 [f]
$2p3d\,{}^1P^\circ$	64.284 ± 0.01 [h]		64.172 [b]

[a]Based on an experimental energy for $2p^2\,{}^3P$ of (59.6722 ± 0.0002) eV from Ref. 10.
[b]Reference 15.
[c]Reference 22.
[d]Based on a theoretical energy for $2s2p\,{}^3P^\circ$ of 58.308 eV from Ref. 20.
[e]Reference 17.
[f]Reference 18.
[g]Reference 25.
[h]Based on a theoretical energy for $2p3d\,{}^1D^\circ$ of 63.664 eV from Ref. 22.

length with many of the satellite lines of C v and some of those of B iv. A solution to this problem might be to look for transitions between the doubly excited terms which lie at longer wavelengths.[22]

IV. CONCLUSION

The main conclusion is that calculations of energies in the two-electron system with both electrons excited are generally in good agreement with experiment. Apart from the observations of the $^1P^\circ$ series by Madden and Codling,[2] and of the $2p^2\,^3P$ term in emission,[10] these are the first accurate measurements of the term values. In Table IV we give the resulting energies of the terms observed.

The measured lifetimes agree well with the calculated transition probabilities of Drake and coworkers.[19,22] The observation that the radiative transition at 2577 Å from $pp23-{}^3D$ is very strong and its measured mean life suggest that its autoionization rate is very small, closer to the value of Cooper *et al.*[17] than to that of Altick and Moore.[18] The autoionization width of the $2s2p\,^3P$ term is found to be in good agreement with theory for both He i and Li ii.[20]

The beam-foil technique appears to very efficiently excite these doubly excited states in comparison to other light sources such as hollow cathodes and spark discharges. This has already been noted for Li i[12] and appears to hold, in general, for other ions. However, only transitions from states with very small autoionization widths ($<10^{-4}$ eV) have been observed.

ACKNOWLEDGMENTS

We are grateful to Professor A. Dalgarno, Professor G. W. F. Drake, and Professor E. Holøien for their suggestions and the communications of their results in advance of publication. We gratefully acknowledge Professor Edlén's kindness in communicating his unpublished results and allowing us to publish them.

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