

Lifetimes of Some Doubly Excited Levels in Neutral Helium[†]

H. G. Berry,* I. Martinson, L. J. Curtis,† and L. Lundin

Research Institute for Physics, 104 05 Stockholm, Sweden

(Received 3 November 1970)

We have searched for radiative transitions from doubly excited levels in He I, using the beam-foil method. The spectra showed the $1s2p\ ^3P-2p^2\ ^3P$ transition at 320.4 Å and some weaker lines, which are also interpreted as transitions from the doubly excited system. Radiative lifetimes were measured for three lines. Our value for the $2p^2\ ^3P$ level, 0.080 ± 0.007 nsec, is in agreement with the recent theoretical value of 0.0803 nsec. The possibilities of observing transitions from doubly excited levels in the He I isoelectronic sequence are briefly discussed.

INTRODUCTION

Evidence of doubly excited states in neutral helium was first obtained from studies of the arc spectrum of helium^{1,2} and from energy-loss measurements in electron-helium collisions.³ Compton and Boyce¹ and Kruger² reported unidentified spectral lines at 309.04 and 320.38 Å. The former remained unclassified, whereas Kruger tentatively assigned the latter to the $1s2p\ ^3P-2p^2\ ^3P$ transition in He I. This identification was later supported by Wu's calculations⁴ of auto-ionization probabilities for various doubly excited terms in helium, of which those of the type $2pnp\ ^1P$, 3P , and $2pnd\ ^1D$, 3D are not expected to auto-ionize via Coulomb interaction.

In recent years, the auto-ionizing doubly excited He I levels have been the subject of many experimental and theoretical investigations. The review article by Fano⁵ gives a detailed list of references. Using synchrotron radiation to excite neutral helium, Madden and Codling^{6,7} observed four Rydberg series in the doubly excited 1P system. Two of the series converged to the $n=2$ limit of He⁺ and were described by Cooper *et al.*⁸ as $sp2n \pm (n \geq 3)$, being symmetrized mixtures of the $2snp$ and $2pns$ series

members. Several of these and other doubly excited levels have also been observed as resonances in electron-helium⁹⁻¹³ and ion-helium¹⁴ collisions. Most of the doubly excited levels observed so far can autoionize to the continua above the $1s\ ^2S$ ground state of He⁺. From their Fano-type line profiles,¹⁵ Madden and Codling⁷ were able to deduce the auto-ionization probabilities for the $2s2p\ ^1P$ and $sp23 + ^1P$ levels, obtaining good agreement with theory.¹⁶ The radiative deexcitation probabilities for several auto-ionizing 1P and 3P states have been calculated by Knox and Rudge¹⁷ and Dickinson and Rudge.¹⁸

Experimental and theoretical results for the non-auto-ionizing doubly excited He I states are not as numerous. The energy of the $2p^2\ ^3P$ level has been calculated by, among others, Holøien,¹⁹ Midtdal,²⁰ and Drake and Dalgarno.²¹ Their eigenvalues give further support to the assignment of the 320-Å line. Drake and Dalgarno have also calculated the energy of the $2p3p\ ^1P$ level and the lifetimes of both these levels. They identify the 309-Å line, observed by Compton and Boyce,¹ as the $1s3p\ ^1P-2p3p\ ^1P$ transition. The need for further experimental studies of such exactly quantized doubly excited levels has been emphasized by Holøien.²² This article de-

scribes an investigation of these levels, using the beam-foil technique.

EXPERIMENT

We accelerated beams of He^+ to energies between 200 and 500 keV in a 2-MV Van de Graaff accelerator and directed them through a thin exciter foil (typically $10 \mu\text{g}/\text{cm}^2$). Initially only carbon foils were used, but we found that by evaporating a very thin metal layer (e.g., Al, Cu, or Ag) on the downstream side of the foil, the light output increased by a factor of 2 or more. The radiation emitted by the foil-excited He atoms and ions was dispersed with a 3-m grazing incidence spectrometer of the Siegbahn type, equipped with a 540 1/mm grating. Reference 23 presents a more detailed description of the experimental arrangement. The photons were counted in a single-channel analyzer and the lifetimes were measured by recording the intensity decrease of the spectral lines with the distance from the foil. Data were accumulated for a fixed amount of charge, collected in a Faraday cup, and 0.1 dark counts were subtracted for each second of data accumulation. To check the experimental accuracy, the decay of the strong 303.8-Å line in He II ($1s^2S-2p^2P$) was measured after most measurements of the doubly excited levels, using the same foils and same beam particle energies. The results for the lifetime of the $2p^2P$ level were within 5% of the theoretical value 0.0998 nsec.²⁴

RESULTS

Wavelengths

Figure 1 shows a partial spectral scan around the 303.8-Å line. The linewidths of 1 Å are essentially instrumental in origin, the line broadening due to the scattering in the foil being about 0.1 Å at the beam energies used.

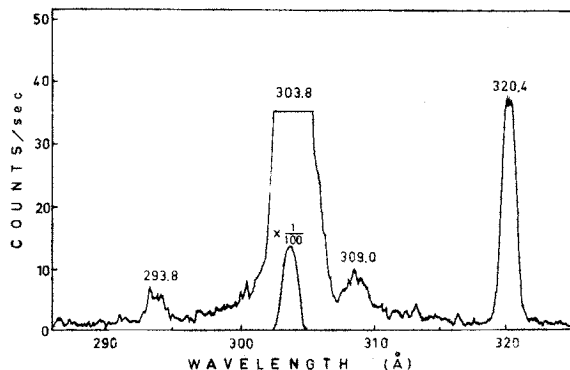


FIG. 1. Beam-foil spectrum of helium between 285 and 325 Å. The incoming beam-particle energy was 240 keV. The photons were detected with a Bendix channel-tron movable on the Rowland circle. The detection system was designed by Dr. H. Oona, University of Arizona.

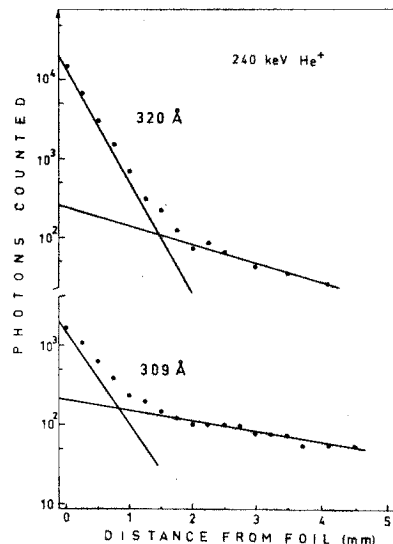


FIG. 2. Decays of the $2p^2\ ^3P$ (320 Å) and $2p3p\ ^1P$ (309 Å) levels in He I.

The $1s2p\ ^3P-2p^2\ ^3P$ transition (320 Å) was clearly seen in all spectra. The intensity ratio 320 Å/303 Å was 0.014 at 320 keV and decreased to 0.009 at 500-keV incoming particle energy, consistent with the fact that the lines belong to different ionization stages. Other weak lines were also observed, and are given in Table I. Our experimental wavelength uncertainties are less than 0.5 Å, except for the weakest lines, which are accurate to within ± 1 Å. These wavelength uncertainties together with the limited knowledge of the non-auto-ionizing doubly excited levels makes identifications difficult. However, we present in Table I some tentative classifications, using the energies calculated in Refs. 17, 18, 21, and the unpublished $2p3p\ ^3P$ eigenvalue of -1.1356256 Ry.²² The $sp23-^1P$ terms are expected to be nearly stable against auto-ionization⁸ and their radiative deexcitations to singly excited 1S terms could be the origin of some of the weak lines in our spectra. The line at 309.0 Å in the beam-foil spectra is probably the $1s3p\ ^1P-2p3p\ ^1P$ transition. The corresponding triplet transition $1s3p\ ^3P-2p3p\ ^3P$ has a theoretical wavelength of 305.8 Å.²² We noticed an unresolved wing at 306 Å in the strong 303-Å line, which could be this transition. We have tentatively classified the line observed at 293.8 Å as the $1s2s\ ^1S-sp23-^1P$ transition. Also considered were the $1s2p\ ^1P-2p3p\ ^1P$ and $1s2p\ ^3P-2p3p\ ^3P$ transitions, which have calculated wavelengths of 295.2 and 291.1 Å, respectively,^{21,22} but these values differ too much from the observed wavelength. However, we observed a very weak line at 295.2 Å, whereas no transitions could be observed around 291 Å. It is interesting that transitions from the $2p3p\ ^3P$ level are not more prominent in our spectra, but it is pos-

TABLE I. Transitions and radiative lifetimes of doubly excited levels in He I.

| Observed wavelength (Å) ^a | Transition | Calculated wavelength (Å) | Lifetime of upper level (nsec) | |
|---|--|------------------------------|--------------------------------|---------------------|
| | | | This work | Theory |
| 285 ± 1 | 1s2s ³ S-sp23- ³ P (?) | 285.3 ^b | | |
| 293.8 | 1s2s ¹ S-sp23- ¹ P | 294.0 ^c | 0.116 ± 0.020 | 0.36 ^c |
| 295.2 | 1s2p ¹ P-2p3p ¹ P | 295.2 ^d | | |
| 306 ± 1 | 1s3p ³ P-2p3p ³ P (?) | 305.8 ^e | | |
| 309.0 | 1s3p ¹ P-2p3p ¹ P | 309.0 ^d | 0.105 ± 0.015 | 0.0975 ^d |
| 311.0 | 1s3s ¹ S-sp23- ¹ P | 311.0 ^c | | |
| 320.4 ± 0.3 | 1s2p ³ P-2p ² 3P | 320.3 ^d | 0.080 ± 0.007 | 0.0803 ^d |

^aThe transitions listed here appeared reproducibly in all spectral scans at 240 keV, and were not attributed to instrumental reflections or ghosts. The wavelength uncertainties are ±0.5 Å, unless otherwise noted.

^bReference 17.

^cReference 18.

^dReference 21.

^eReference 22.

sible that the lowest non-auto-ionizing singlet and triplet states $2p3p^1P$ and $2p^23P$ are preferentially populated at the beam-foil interaction. In Table I, we also give preliminary classifications of a few additional lines, also observed in the beam-foil spectra. Kruger's tables² include also a line at 357.5 Å. We searched for this line but found no indications of it in our spectra.

Lifetimes

We were able to follow the intensity decays of the spectral lines at 320, 309, and 294 Å. The results are given in Table I. Figure 2 shows two examples of decay curves. The decay of the $2p^23P$ level (320 Å) was measured at 240 and 500 keV and the value 0.080 ± 0.007 nsec is the mean value of five measurements. The inverse of this value, $(1.25 \pm 0.11) \times 10^{10} \text{ sec}^{-1}$, is in excellent agreement with the sum of probabilities for transitions from $2p^23P$ to $1s2p$, $3p$, and $4p^3P$, $1.245 \times 10^{10} \text{ sec}^{-1}$, calculated by Drake and Dalgarno.²¹ The $2p^23P$ level may also decay to the auto-ionizing level $2s2p^3P$, at a wavelength of 9400 ± 20 Å, but this has not been observed. The close agreement between this work and theory suggests that the probability for the $2s2p^3P-2p^23P$ transition is comparatively low. The decay curve in Fig. 2 indicates that the 320-Å line has a weak cascade contribution, corresponding to a lifetime of approximately 1 nsec. The cascading may be due to transitions from the $2p3d^3D$ level, the energy of which has been calculated by Becker and Dahler.²⁵ Possible radiative transitions $2p^23P-2p3d^3D$ should have a wavelength of about 3000 Å. In a beam-foil experiment, using 83-keV He⁺ from an isotope separator, a few weak lines were observed,²⁶ which may originate from doubly excited D terms. The decay curves for the 309-Å line infer a lifetime of 0.105 ± 0.015 nsec for the $2p3p^1P$ level, which agrees with the theoretical value of 0.0975 nsec.²¹ The effects of cascading were more pronounced, with a cascade lifetime of 1.2 nsec, the origin of

which is unknown. Measurements of the 294-Å transition yielded a lifetime of 0.116 ± 0.020 nsec, which differs from the radiative lifetime of 0.36 nsec for the $sp23-^1P$ level, estimated by Dickinson and Rudge.¹⁸ However, if the auto-ionization channel is not completely closed, a shorter experimental lifetime is plausible.

He I ISOELECTRONIC SEQUENCE

The $1s2p^3P-2p^23P$ transition has been observed in Be III (78.662 Å) by Goldsmith²⁷ and in B IV, C V, and O VII by Edlén and Tyrén.²⁸ The experimental wavelengths agree very well with Drake and Dalgarno's calculations for the He I isoelectronic sequence.²¹ The Be III transition appears also in spark spectra, taken with our grazing incidence spectrograph.²⁹ Goldsmith has identified radiative transitions from the auto-ionizing $2s2p^3P$ and $2p^21D$ levels in Be III. In the beam-foil spectra of helium, no He I transitions from these levels could be observed. This is hardly surprising, however, because in He I their auto-ionization probabilities are expected to be orders of magnitude higher than the radiative decay constants.

Work is in progress to extend the beam-foil studies of doubly excited terms to higher members of the He I isoelectronic sequence.

ACKNOWLEDGMENTS

We are greatly indebted to Professor M. Siegbahn for his stimulating interest and for placing his excellent grazing incidence spectrometer at our disposal. We are also grateful to Professor W. S. Bickel, Dr. H. Oona, and Dr. J. Bromander for several important contributions and to J. Hilke for expert technical advice. Professor B. Edlén and Professor E. Holóien have kindly communicated to us their unpublished material on doubly excited states, and we have also benefited from correspondence with Dr. R. D. Cowan and Dr. H. O. Dickinson.

†Work supported by the Swedish Natural Science Research Council (NFR).

*Present address: Faculté des Sciences, Université de Lyon, 69 Villeurbanne, France.

[‡]On leave of absence from the University of Toledo, Toledo, Ohio 43606.

¹K. T. Compton and J. C. Boyce, *J. Franklin Inst.* **205**, 497 (1928).

²P. G. Kruger, *Phys. Rev.* **36**, 855 (1930).

³R. Whiddington and H. Priestley, *Proc. Roy. Soc. (London)* **A145**, 462 (1934).

⁴T. Y. Wu, *Phys. Rev.* **66**, 291 (1944).

⁵U. Fano, in *Atomic Physics*, edited by V. W. Hughes, B. Bederson, V. W. Cohen, and F. M. J. Pichanick (Plenum, New York, 1969), p. 209.

⁶R. P. Madden and K. Codling, *Phys. Rev. Letters* **10**, 516 (1963).

⁷R. P. Madden and K. Codling, *Astrophys. J.* **141**, 364 (1965).

⁸J. W. Cooper, U. Fano, and F. Prats, *Phys. Rev. Letters* **10**, 518 (1963).

⁹C. E. Kuyatt, J. A. Simpson, and S. R. Mielczarek, *Phys. Rev.* **138**, A385 (1965).

¹⁰P. D. Burrow and G. J. Schulz, *Phys. Rev. Letters* **22**, 1271 (1969).

¹¹J. T. Grissom, R. N. Compton, and W. R. Garrett, *Phys. Letters* **30A**, 117 (1969).

¹²N. Oda, F. Nishimura, and S. Tahira, *Phys. Rev. Letters* **24**, 42 (1970).

¹³S. M. Silverman and E. N. Lassetre, *J. Chem.*

Phys. **40**, 1265 (1964).

¹⁴M. E. Rudd, *Phys. Rev. Letters* **15**, 580 (1965).

¹⁵U. Fano, *Phys. Rev.* **124**, 1866 (1961).

¹⁶P. G. Burke, D. D. McVicar, and K. Smith, *Phys. Rev. Letters* **11**, 559 (1963).

¹⁷H. O. Knox and M. R. H. Rudge, *J. Phys. B* **2**, 521 (1969).

¹⁸H. O. Dickinson and M. R. H. Rudge, *J. Phys. B* **3**, 1284 (1970).

¹⁹E. Holøien, *J. Chem. Phys.* **29**, 676 (1958); and *Phys. Norvegica* **1**, 53 (1961).

²⁰J. Midtdal, *Phys. Rev.* **138**, A1010 (1965).

²¹G. W. F. Drake and A. Dalgarno, *Phys. Rev. A* **1**, 1325 (1970).

²²E. Holøien, *Nucl. Instr. Methods* **90**, 229 (1970); and (private communication).

²³L. Lundin, H. Oona, W. S. Bickel, and I. Martinson, *Physica Scripta* **2**, 213 (1970).

²⁴W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities*, U. S. Natl. Bur. Std. National Reference Data Series-4 (U. S. GPO, Washington, D. C. 1966), Vol. I.

²⁵P. M. Becker and J. S. Dahler, *Phys. Rev.* **136**, A73 (1964).

²⁶J. Bromander and H. G. Berry (unpublished).

²⁷S. Goldsmith, *J. Phys. B* **2**, 1075 (1969).

²⁸B. Edlén and F. Tyrén, *Nature* **143**, 940 (1939); and (unpublished).

²⁹M. Siegbahn and L. Lundin (unpublished).