Accurate Wavelength Measurement of the $1s2p\ ^3P^\circ$-$2p^2\ ^3P$ Transition in ^{4}He I

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(Received 28 June 1971)

An accurate spectroscopic measurement has yielded 320.2926 ± 0.0010 Å for the wavelength of the $1s2p\ ^3P^\circ$-$2p^2\ ^3P$ transition in ^{4}He I. Combining this wavelength with the well-known position of the $1s2p$ term gives an experimental value of $481\,301.5 \pm 1.2$ cm^{-1} for the position of the doubly excited $2p^2\ ^3P$ term relative to the ground $1s^2\ ^1S$. The new measurement is in excellent agreement with Aashamar's theoretical calculation of 481 301.6 cm^{-1} for the energy of the term.

Numerous recent papers have been devoted to experimental and theoretical studies of multiply excited energy states in atoms. Doubly excited states in the He I isoelectronic sequence have received particular attention because this two-electron sequence is the most amenable to exact theoretical treatment. A review of recent developments, including a comprehensive bibliography, has been provided by Holøien.[1]

The doubly excited states of the helium atom, all of which lie embedded in the He I continuum, can be divided into two general groups according to their stability, in the nonrelativistic approximation, against autoionization through Coulomb interaction with the continuum. Those weakly quantized states of ^{4}He I that are short lived with respect to autoionization have been the subject of many recent investigations,[2] both theoretical and experimental. Accurate spectroscopic measurements of energies for the exactly quantized bound states, however, are almost nonexistent. These states have no interaction with the continuum owing to the conservation requirements on parity and angular momenta, and sharp-line transitions from them to lower one-electron excitation states should be observable. If relativistic interactions are taken into account, autoionization becomes possible even for the exactly quantized states, but the autoionization lifetimes are still several orders of magnitude longer than the mean radiative lifetimes of the allowed electric dipole transitions.[1] Until recently, the only transition from one of these doubly excited levels definitely identified spectroscopically in emission was the line measured at 320.392 Å by Kruger.[3] Kruger's interpretation of this line as the He I $1s2p\ ^3P^\circ$-$2p^2\ ^3P$ transition was later confirmed by Wu[4] on the basis of persuasive theoretical arguments. Berry *et al.*[5] have now observed this line and six others in a beam-foil experiment and have assigned tentative identifications to all of them on the basis of existing calculations. The wavelength uncertainties given for the lines measured in their experiment range from ±0.3 to ±1 Å.

Among the nonautoionizing doubly excited states in helium, the $2p^2\ ^3P$ term is the lowest and has received extensive theoretical attention by self-consistent field, variational, and variational-perturbational techniques.[6-11] The most accurate and inclusive calculation is that by Aashamar,[11] who took into account mass-polarization, relativistic, and radiative contributions in calculating the total energy of the $2p^2\ ^3P_0$ level by means of Hylleraas-Scherr-Knight (HSK) variational-perturbational wave functions. He found the theoretical fine-structure splitting to be

$$E_{\mathrm{th}}(^3P_0) - E_{\mathrm{th}}(^3P_1) = -1.275\,963\,4\ \mathrm{cm}^{-1}$$

and

$$E_{\mathrm{th}}(^3P_0) - E_{\mathrm{th}}(^3P_2) = -3.170\,195\,2\ \mathrm{cm}^{-1}.$$

This implies a normal term whose center of gravity is 2.19 cm^{-1} higher than the predicted lowest level $^3P_0$. All other theoretical calculations of the $2p^2\ ^3P$ term have been in the nonrelativistic approximation, and therefore do not yield the fine structure; but one of them, by Bhatia,[10] does include the mass-polarization correction. Bhatia[10] has tabulated several of the more recent calculated values for the wave number of the $1s2p\ ^3P^\circ$-$2p^2\ ^3P$ transition. The wavelengths predicted for this line in ^{4}He I range from 320.288 to 320.293 Å when the theoretical energies of the $2p^2\ ^3P$ term as calculated by different authors are combined with the well-known energy[12] of the $1s2p\ ^3P^\circ$ term. This gives a discrepancy between observed[3] and theoretical energies of the $2p^2\ ^3P$ term of about 100 cm^{-1}. A discrepancy of this size cannot be explained on theoretical grounds. The investigation reported here was undertaken to obtain a new spectroscopic measurement of the transition and to provide a more accurate comparison between theory and experiment.

All spectrograms were obtained by use of the National Bureau of Standards 10.7-m vacuum Eagle spectrograph, equipped with a 175×100 mm^2 concave grating having 1200 grooves per mm. The reciprocal dispersion is 0.78 Å/mm in the first order and the resolving power in the higher orders is about 300 000. The light source used was a helium discharge excited by a 13.56-MHz power supply. The gas pressure in the source is estimated to have been about 0.2 Torr.

The exposure times necessary to register the He I λ320 line with sufficient intensity on Kodak Special Plates, type 101-05, ranged from 5 to 7 h. Although the line was recorded in the first four grating orders, the measurements reported here are from spectrograms on which the line appears in the second and third orders. The first-order exposure lacks standard comparison lines, and the line appears to be a blend on the fourth-order exposure.

First-order lines of Ar II from separate exposure to a low-pressure argon discharge and first-order lines of O I, N I, N II, H I, and C II excited simultaneously as impurities in the helium discharge served as internal standards of wavelength. The values for the standard wavelengths used, nearly all of which have estimated uncertainties of less than ±0.0007 Å, were taken from an unpublished list compiled by Kaufman.[13]

A summary of our measurements is contained in Table I. The intensities given in column 5 of this table are visual estimates of the ratio of the intensity of the λ320 line to that of the weakest line easily discernible on the same spectrogram. The width of the λ320 line on our plates was essentially the instrumental width and about the same as that of other lines of comparable intensity on the plates, but no precision measurements of linewidths were attempted.

With one exception, each plate was measured more than once, on different measuring engines. Measurement of several other sharp lines having accurately known wavelengths, such as He II λ257, in the various grating orders gave no indication of an order shift. Any such shift of order coincidence must amount to less than ±0.001 Å. The standard deviations listed in Table I for each plate reduction were calculated from the residuals of the fit of the first-order standard lines to the least-squares polynomials used to represent wavelength as a function of distance along the individual spectrograms. The corresponding standard deviations appropriate to the He I λ320 line itself are found by dividing these figures by the grating order in which the line was measured. The weighted average of our measurements is 320.2926 ± 0.0007 Å (standard deviation). The error estimate associated with our final adopted value for the ^{4}He I line, 320.2926 ± 0.0010 Å, allows for the possibility of a small error introduced by using the method of overlapping orders and for the uncertainty in the absolute values of the wavelength standards used. Adding the wave number of the line to the known[12] position of the $1s2p\,^3P^\circ$ term, 169 087.01 ± 0.15 cm^{-1}, gives an experimental value of 481 301.5 ± 1.2 cm^{-1} for the energy of the doubly excited $2p^2\,^3P$ term relative to the ground $1s^2\,^3S$.

In comparing this measurement with theory, we confine our attention to Aashamar's relativistic calculation of ^{4}He $2p^2\,^3P$. The most accurate comparison is obtained by combining this calculation with the known experimental energy[12] of the $1s2p$ term to predict the wavelength of the transition between the two.[14] Since the fine structure

TABLE I. Summary of measurements of the wavelength of the $1s2p\,^3P^\circ$–$2p^2\,^3P$ transition in ^{4}He I.

Plate number	Grating order	Number of standards	Standard deviation (Å)	Intensity	Wavelength (Å)	Weight
X-290	3	28	0.0010	10	320.2926	1
	3	39	0.0013	10	320.2925	1
X-292	2	48	0.0009	15	320.2912	1
	2	48	0.0010	15	320.2930	1
	2	49	0.0006	15	320.2926	1
X-295	3	50	0.0009	25	320.2927	2
X-291	3	28	0.0009	5	320.2938	1
	3	31	0.0020	5	320.2922	1
		Average 320.2926 ±0.0007 Å (standard deviation)				

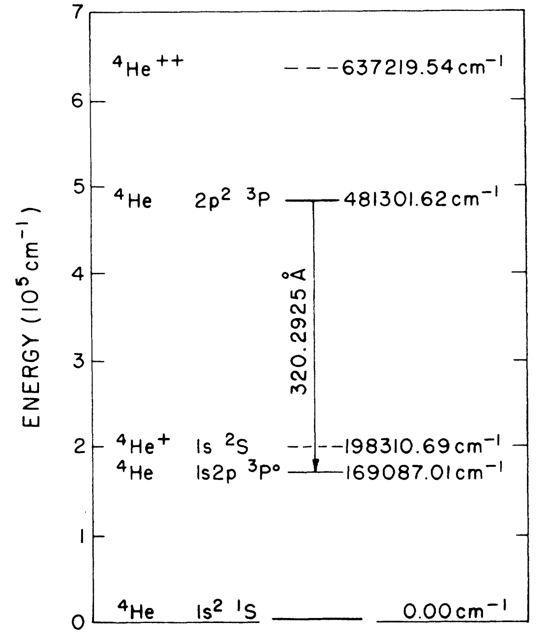

FIG. 1. Partial energy-level diagram for ^{4}He showing the theoretical position of the doubly excited $2p^2\,^3P$ term and the predicted wavelength of the $1s2p\,^3P°$-$2p^2\,^3P$ transition.

of the λ320 line could not be resolved in our experiment, we assume that the measured wavelength represents the transition between the centers of gravity of the two ^3P terms. The important relativistic correction included in the calculation of the theoretical position of the $2p^2$ term amounts to about 6 cm^{-1}, or six times the experimental uncertainty of our measurement.

A partial energy-level diagram for ^{4}He is shown in Fig. 1. The position of the $2p^2$ term above the ground $1s^2\,^1S_0$ level was obtained by subtracting Aashamar's total energy for the $2p^2$ term from the total energy of the ^{4}He $1s^2\,^1S_0$ ground level, $-637\,219.54$ cm^{-1}. This latter energy is taken as the sum of the ionization potentials of the ^{4}He atom as calculated by Aashamar[11] and of the ^{4}He$^+$ ion as calculated by Garcia and Mack.[16] The center of gravity of the $1s2p\,^3P°$ term is an experimental one,[12] accurate to ±0.15 cm^{-1} in absolute value.

As seen from Fig. 1, Aashamar's theoretical calculation of the ^{4}He $2p^2\,^3P$ term agrees with our new experimental determination to well within 1 cm^{-1}, which is the wave number uncertainty associated with an error of 0.001 Å at this wavelength. The disturbing 100-cm^{-1} discrepancy indicated by the earlier measurement of this line, which prompted the present investigation, appears to have been primarily a result of the unavailability of accurate wavelengths to serve as comparison standards at the time of that measurement.

One of us (J.L.T.) has observed the He I λ320 line in connection with the optical alignment of the new National Bureau of Standards 10.7-m grazing-incidence vacuum spectrograph. It is hoped that the increased speed of this instrument, in combination with an improved helium light source, will permit accurate measurement of transitions from additional doubly excited nonautoionizing states in ^{4}He.

We are indebted to Wm. C. Martin for inspiring this investigation and for frequent discussions of the problem. Conversations with J. W. Cooper were helpful in locating the source of the discrepancy discussed in Ref. 14.

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[1] E. Holøien, Nucl. Instrum. Methods 90, 229 (1970).

[2] See, for example, the review article by U. 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.

[3] P. G. Kruger, Phys. Rev. 36, 855 (1930).

[4] T.-Y. Wu, Phys. Rev. 66, 291 (1944).

[5] H. G. Berry, I. Martinson, L. J. Curtis, and L. Lundin, Phys. Rev. A 3, 1934 (1971).

[6] W. Wilson, Phys. Rev. 48, 536 (1935).

[7] A. F. Stevenson, Phys. Rev. 56, 586 (1939).

[8] G. W. F. Drake and A. Dalgarno, Phys. Rev. A 1, 1325 (1970).

[9] E. Holøien, in *Proceedings of the Fourth International Conference on the Physics of Electronic and Atomic Collisions* (Science Bookcrafters, New York, 1965), p. 6.

[10] A. K. Bhatia, Phys. Rev. A 2, 1667 (1970).

[11] K. Aashamar, Nucl. Instrum. Methods 90, 263 (1970).

[12] W. C. Martin, J. Res. Nat. Bur. Stand., Sect. A 64, 19 (1960).

[13] V. Kaufman, unpublished.

[14] In predicting the wave number of the $1s2p\,^3P°$-$2p^2\,^3P$ transition, we found a disagreement of 3.69 cm^{-1}, depending upon whether we used the experimental value for the $1s2p$ term or the theoretical value as given by Aashamar (Ref. 11) and Bhatia (Ref. 10). The latter authors arithmetically reconstructed the *total* energy, $E = -4.266\,556\,218 R_{\text{He}}$, of the $1s2p$ term from the published calculation of the $1s2p$ *ionization* energy by B. Schiff, H. Lifson, C. L. Pekeris, and P. Rabinowitz, Phys. Rev. 140, A1104 (1965). In calculating the total energy, however, they neglected to take into account the radiative (Lamb-shift) correction of the term. Since the ionization energy of the $1s2p\,^3P_1$ level as calculated by Schiff *et al.* differs from the experimental value by only 0.03 cm^{-1}, the Lamb-shift correction to the total energy stated above must be very close to the Lamb shift of the $1s\,^2S$ level in the helium ion, or 3.53

cm^{-1}. The predicted wave numbers tabulated by Bhatia in Table III of his paper (Ref. 10) should, therefore, be reduced by this amount.

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[15] Schiff, Lifson, Pekeris, and Rabinowitz, see Ref. 14.

[16] J. D. Garcia and J. E. Mack, J. Opt. Soc. Amer. 55, 654 (1965).

Zero-Field Intensity Oscillations Following Impulsive Excitation of Hydrogen*

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(Received 20 April 1971)

In a beam-foil experiment, we have observed zero-field oscillations of approximately 25% in the intensity of H_β radiation polarized parallel to the beam. An unmodulated decay is observed when a signal proportional to the total intensity is examined. Our observations substantiate Macek's theory of coherent decay from fine-structure levels.

The first evidence that the decay of radiation from an excited state might show an oscillatory time dependence under zero-field conditions was provided in an experiment by Bashkin and Beauchemin.[1] They observed the decay of helium ions impulsively excited in a carbon foil. Macek[2] suggested that such oscillations could be caused by an interference between fine-structure levels, under completely field-free conditions. Subsequently, Andrä[3] observed zero-field oscillations in helium and for $n=3, 4$ in hydrogen. In the case of H_β the data could be decomposed into frequencies 1390 and 440 MHz and possibly 1500 MHz. These frequencies represent the fine-structure separations of $p_{1/2}$-$p_{3/2}$ (1371 MHz, $d_{3/2}$-$d_{5/2}$ (457 MHz), and a possible mixture of $s_{1/2}$-$d_{3/2}$ (1233 MHz and $s_{1/2}$-$d_{5/2}$ (1690 MHz). Sellin, Biggerstaff, and Griffin[4] reported on a similar experiment on H_β and H_γ, and essentially observed no oscillations within the limits set by their apparatus. Andrä's work was performed in the earth's magnetic field where the motional electric field experienced by the moving atom ($\vec{v}\times\vec{B}$) is approximately 2 V/cm. Sellin, Biggerstaff, and Griffin performed their experiment in a chamber which was surrounded by Helmholz coils which reduced the magnetic field to 50 mG. We have successfully tested two facets of Macek's interpretation of zero-field oscillations.

All present observations were made with carbon foils of nominal thickness 10 $\mu\mathrm{g/cm}^2$.[5] The 130-keV proton beam was collimated by two $\frac{1}{8}$-in. apertures, the first being located in the beam pipe and the second directly in front of the foil. These apertures were separated by an initial distance of 13 cm (see Fig. 1). Subsequent to excitation, observations were made on the 4861 Å H_β emission from the hydrogen component of the beam, whose velocity was 5×10^8 cm/sec.[6] Beam currents of 15–20 μA were collected in the Faraday cup.

The foil and decay region were enclosed within a magnetic shield. The component of the magnetic field normal to the beam was measured to be 37.6 mG. The motional electric field experienced by an atom in the beam was thus 0.19 V/cm.

The radiation was detected by a 1-m, normal incidence, McPherson Model No. 225 monochromator with a reciprocal dispersion of 16 Å/mm, using 1-mm slits and an uncooled EMI-6256S photomultiplier. The signal was processed by a conventional counting system (Fig. 1), with the scaler gated from a current integrator set to stop the scaler when a preset charge was accumulated in the Faraday cup (usually 12×10^{-4} C). A rotatable polarizer (type HN22 Polaroid) was placed close to the entrance slit of the mono-

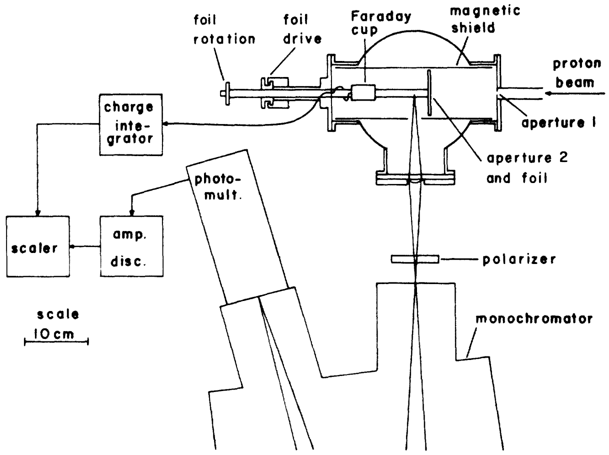

FIG. 1. Foil drive mechanism and optical detection system.

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