Energy and lifetime measurements in heliumlike and lithiumlike phosphorus*

Ph. Deschepper, † P. Lebrun, L. Palffy, ‡ and P. Pellegrin

Institut de Physique Corpusculaire, 2, Chemin du Cyclotron, 1348 Louvain-la-Neuve, Belgium (Received 18 January 1982)

The x-ray energies in the transitions to the ground state from the 2^3P_2 and 2^3S_1 levels in heliumlike phosphorus and from the $2^4P_{5/2}$ level in lithiumlike phosphorus have been determined by using a Doppler-tuned x-ray spectrometer. The experimental results $E(2^3P_2-1^1S_0)=(2142.3\pm1.2)$ eV, $E(2^3S_1-1^1S_0)=(2126.7\pm1.2)$ eV, and $E(^4P_{5/2}-^2S_{1/2})=(2112.3\pm1.2)$ eV are in good agreement with theoretical predictions. The mean life of the $2^4P_{5/2}$ and 2^3P_2 states have also been measured by the beam-foil time-of-flight method. The experimental values $\tau(2^3P_2)=(3.6\pm0.1)\times10^{-9}$ s and $\tau(2^4P_{5/2})=(1.2\pm0.1)\times10^{-9}$ s are in agreement with theoretical predictions.

I. INTRODUCTION

For several years the physics of highly ionized atoms has received much interest from both experimentalists and theoreticians. The development of heavy-ion accelerators permits production of high-Z few-electron atoms which can be investigated by means of the well-known beam-foil technique. The information gathered on atomic structure and radiative or autoionizing decays is exploited in many fields such as astrophysics, plasma, and condensed-matter physics. New methods have also been developed in the field of relativistic many-electron theory $^{1-5}$ and quantum electrodynamics. 6,7

One of the outstanding problems is the study of the metastable states of He-like and Li-like atoms. The strict LS coupling scheme, which forbids completely singlet-triplet intercombination transitions, is a simplified view of the real situation. In fact, this scheme never holds exactly, but in light atoms the violation is so small that the observation of such lines is very difficult.⁸ The situation completely changes in atoms with high nuclear charge. The electrons are closer to each other and move at higher velocities, giving rise to strong spin-orbit interactions. As a consequence, spin-flip transitions become possible between singlet and triplet systems with rates growing with high powers of the nuclear charge. Figure 1 represents the level structure of He-like ions; the principal decay modes are indicated. Owing to the spin-orbit interaction, the $2^{3}P_{1}$ state is no longer a pure triplet state; it is admixed to some amount with the $2^{1}P_{1}$ singlet state and decays to the ground state with a fast E1 transition. This is the first observed intercombination line in beam-foil spectroscopy and was performed by Sellin et al.9 in He-like oxygen and nitrogen. Since this

first experiment, many others have been performed 10,11 on ions up to sulphur (Z=16). Owing to the rapid decrease of the lifetime of the 2^3P_1 level with increasing nuclear charge, such measurements cannot be extended to much higher Z heliumlike ions.

The observation of higher-order multipole radiations was first performed by Marrus and Schmieder

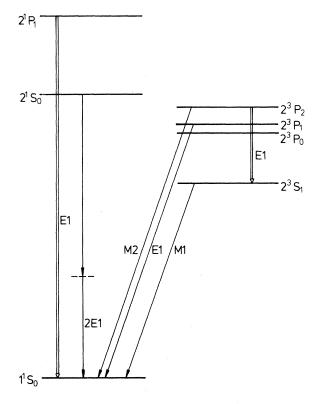


FIG. 1. Low-lying level structure of heliumlike atoms.

in He-like argon. ¹² They investigated, among others, the magnetic quadrupole transition from the 2^3P_2 state to the ground state. It is worthwhile to note that this transition is in competition with an E1 transition to the 2^3S_1 level. Given the different Z dependences of both transition rates $(M2 \propto Z^8; E1 \propto Z)$, the M2 transition is dominant above Z=18: a quite unique case in physics. Other transitions of this type have been observed by several groups. The reader is referred to complete reviews by Cocke, ¹³ Marrus, ¹⁴ and Sellin ¹⁵ for more detailed information.

The 2^3S_1 - 1^1S_0 intercombination line results from a highly forbidden transition: a relativistic M1 transition between states having the same parity but orthogonal spatial wave functions. Given the slow decay rate all along the helium isoelectronic series the investigation of this line is rather difficult. Much theoretical work, stimulated by astrophysics, and experimental work initiated by Marrus and Schmieder has been devoted to the study of this decay.

Interest in the $2^4P_{5/2}$ quartet state in Li-like ions essentially stems from its metastability both under autoionization and radiative decay. Only spin-spin autoionization is permitted, in competition with a M2 transition to the ground state. Extensive studies of this decays have been performed by many authors. ^{15,21}

The aim of this work is to complete this field of atomic physics by measurements performed in our laboratory in He-like and Li-like phosphorus (Z=15) by means of a high-resolution Dopplertuned x-ray spectrometer. Results of the transition

energies of the following forbidden decays are reported: $2^3P_2(M2)1^1S_0$ and $2^3S_1(M1)1^1S_0$ in P^{13+} and $^4P_{5/2}(M2)^2S_{1/2}$ in P^{12+} together with the mean lives of these levels (except the mean life of the very long-lived 2^3S_1 state). In Sec. II we describe the experimental setup, beam transport, stripping chamber, spectrometer, and choice of the critical absorber. Section III is devoted to the measurements and experimental results which will be confronted with theoretical predictions in Sec. IV.

II. EXPERIMENTAL SETUP

P⁵⁺ ions are accelerated up to 87 MeV with the isochronous cyclotron of the University of Louvain-la-neuve (CYCLONE). Typical extracted currents are of the order of 500 nA. At the exit of the cyclotron (Fig. 2) are located ten carbon foils (C1) $100 \,\mu\text{g/cm}^2$ thick fixed on a remote controlled stripper holder. P¹⁴⁺ or P¹³⁺ ions are selected with the steering magnet (M1) and sent to the experimental area. The beam is geometrically defined with two separated adjustable collimators situated upstream of the spectrometer, which contains a movable carbon foil (C2) of $10 \mu g/cm^2$. Depending on the experimental conditions the beam is either integrated in Faraday cup FC2 or is charge analyzed with a second magnet (M2). A particular charge state is then selected and integrated in Faraday cup FC3 or detected with a plastic scintillator viewed by a photomultiplier. Typical currents measured on FC2 are of the order of several nA. A complete charge-state analysis is reported in Ref. 22.

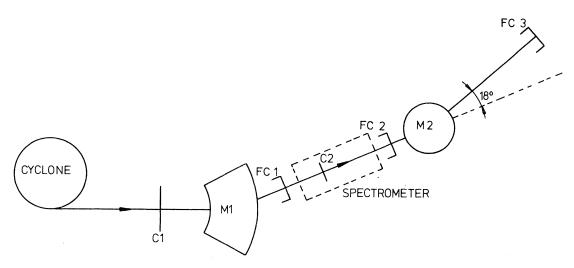


FIG. 2. Scheme of the experimental setup.

The Doppler-tuned x-ray spectrometer consists of a critical absorber and an x-ray counter whose angular position with respect to the beam axis can be varied with a high precision rotator. A detailed description of such a spectrometer is given in Ref. 23. The observed energy E of a photon emitted in direction θ with respect to the beam traveling at velocity β is given by the well-known Doppler formula

$$E = \frac{E_0 (1 - \beta^2)^{1/2}}{1 - \beta \cos \theta} , \qquad (1)$$

where E_0 is the energy of the photon in the rest frame of the emitter. If a suitable absorber having a characteristic absorption edge $E_{\rm abs}$ is placed in front of the detector, the number of detected x rays abruptly diminishes when the detector is rotated at an angle θ_1 such that $E > E_{\rm abs}$. The real energy of the photon is computed from the relation

$$E_0 = \frac{E_{\text{abs}}(1 - \beta \cos \theta_1)}{(1 - \beta_1^2)^{1/2}} \ . \tag{2}$$

An energy measurement thus consists of counting the x rays as a function of the angular position of the detector and to bring the Doppler-shifted energy in coincidence with the known absorption discontinuity. A resolving power of the order of 10^{-4} is easy to achieve. Mechanical details of our spectrometer are shown in Fig. 3. The detection system and movable carbon foils (C2) are located in the same aluminium chamber. The foils are deposited on five metallic supports (with holes of 5 mm diam.) fixed on a multiposition holder. The latter is mounted on a linear carrier moved by means of a synchronus motor and a threaded screw rod (250 turns correspond to a longitudinal displacement of 18.75 mm). Our detector is a usual gas flow (argon-methane) proportional counter. It is provided with Soller slits (angular resolution 0.4°), and an absorption foil of zirconium (4 µm thick). A precision rotator together with a circular track enables one to fix the observation angle with respect to the beam. The theoretical predictions of the x-ray energies we intend to measure lie in the range of 2110-2150 eV. The most convenient absorption edge is the $L_{\rm III}$ discontinuity of zirconium²⁴ because the velocity of the ions is $\beta = 0.0767$. Recommended values of the x-ray wavelengths can be found in many review papers.24-27 Serious discrepancies exist as is illustrated by the variation of the $L_{\rm III}$ edge of zirconium itself:

2220 eV
$$\leq L_{III}(Zr) \leq 2224 \text{ eV}$$
.

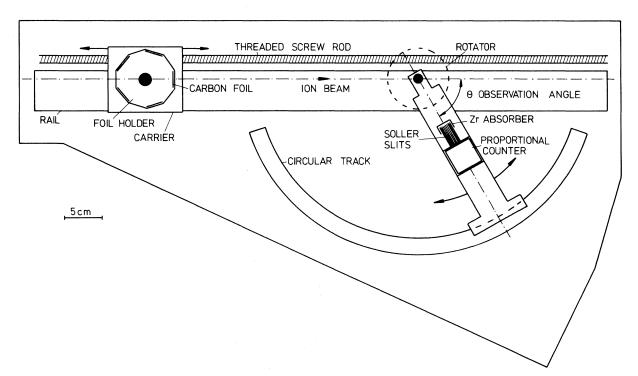


FIG. 3. Diagram of the Doppler-tuned x-ray spectrometer.

In the widely used tables by Bearden,²⁴ one reads $L_{\rm III}(Zr) = 2222.5$ eV. These tables result from an enormous compilation of work based on all published data and on new primary and secondary standards.

Very often, however, the experimental values found in the literature are rather incomplete and inconsistent so that indirect evidence has been used in many instances. Since the publication of these tables, high precision work on x rays have been performed²⁸ giving results which do not agree (within a few eV) with the values proposed by Bearden. In a recent work²⁹ on x-ray spectroscopy in zirconium, the authors, although interested only in the emission lines, also display the measured $L_{\rm III}$ edge. It is well known³⁰ that, in metals, the position of the edge (position of the half maximum) of an emission line coincides with that of the absorption limit. From Ref. 29 we have $L_{\text{III}}(Zr) = 2220.4$ eV in perfect agreement with the only earlier high-precision measurement by Sandström³⁰: $L_{III}(Zr) = 2220.6$ eV. The latter will therefore be adopted in this paper.

III. RESULTS

A. Energy measurements

The 2^3P_2 level is produced by selecting P^{14+} ions with the steering magnet M1 (Fig. 2) and sending them through a second carbon stripper, (C2) located in the chamber of the spectrometer. The foil-detector separation was 8 cm, a compromise between signal and background (essentially due to x rays emitted by P^{13+} ions and part of the two-photon decays in H-like and He-like phosphorus. The measurements are normalized on FC 2 (Fig. 2). The inset of Fig. 4 shows a typical energy spectrum recorded at an angle of $\theta = 63^{\circ}$ and Fig. 4 is the integrated peak as a function of θ . The full width at half maximum (FWHM) of the peak, obtained by differentiation of the curve, is 2 eV.

The $2^4P_{5/2}$ spectrum was obtained in similar conditions. Instead of P^{14+} ions, P^{13+} ions are selected with M1 (Fig. 2); the foil-detector separation was 1.5 cm. Figure 5 represents the counting rate as a function of θ . The less-pronounced step is due to the fact that the production yield of the $2^4P_{5/2}$ level in our conditions is relatively low as compared to the formation of He-like levels appearing as background in this measurement. It is worthwhile to mention that the apparent irregularities in the experimental absorption edge are not due to statistical fluctuations but are systematically reproduced.

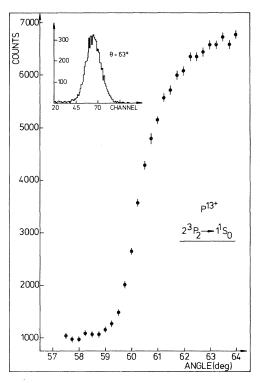


FIG. 4. Spectrum measured with the Doppler tuned x-ray spectrometer for the 2^3P_2 - 1^1S_0 transition. Inset shows a typical energy spectrum recorded with the proportional counter at $\theta = 53^\circ$.

They can be assigned, although in a qualitative manner, in this experiment to the extended (Kronig) fine structure²⁷ of the $L_{\rm III}$ absorption edge of zirconium. The observed curve is, in fact, the mean absorption edge as seen by the $2^4P_{5/2}$ x rays but modulated by the extended fine structure of the edge as seen by the 2^3P_2 background x rays.

The 2^3S_1 x rays are more difficult to observe due to the very long lifetime of the level, 1.4 μ s corresponding to 32.5 m in the laboratory. They are unobservable in the described experimental conditions. To observe them we used only one stripper foil C1. P^{13+} ions produced in the interaction of the accelerated P^{5+} ions with the carbon foil of 100 μ g/cm² are selected with M1 and steered to the spectrometer. Among the ions in excited states, only the very long-lived 2^3S_1 states survive at the position of the detector (foil-detector separation is 21 m). In this way, a very clear spectrum is obtained without any background as is shown by the absence of counting rate below $\theta = 53^\circ$ (Fig. 6) corresponding to the high absorption side of the edge.

In Table I, the measured values of the x-ray energies are displayed. They have been determined by

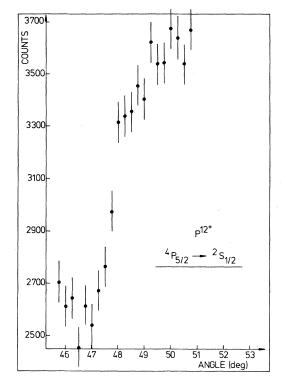


FIG. 5. Measured spectrum of the ${}^4P_{5/2}$ - ${}^1S_{1/2}$ transition.

locating the inflexion point of the absorption edge and using relation (2) together with β =0.0767 corrected for energy losses in the carbon foils. The quoted error of 1.2 eV is a linear combination of (i)

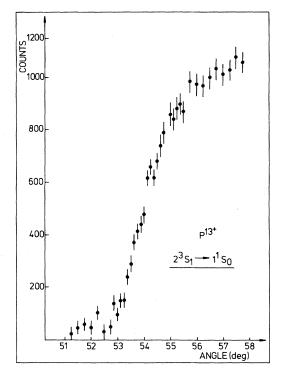


FIG. 6. Measured spectrum of the 2^3S_1 - 1^1S_0 transition.

the uncertainty in the location of the inflexion point: $\pm 0.25^{\circ}$ ($\equiv 0.6$ eV) corresponding to the separation of the experimental points situated at the left and the right of the inflexion point. This experimentally defined error includes the very slight

TABLE I. Comparison of experimental and theoretical values.

Charge state	Transition	Energies (eV)		Lifetimes (ns)	
		Expt.	Theor.	Expt.	Theor.
P ¹²⁺ (Li-like)	$^{4}P_{5/2}$ - $^{2}S_{1/2}$	2112.3±1.2	2111.2ª	1.2±0.1	1.3 ^b
			$2110.6 \pm 0.5^{\circ}$		1.47 ^a
P ¹³⁺ (He-like)	$2^{3}P_{2}-1^{1}S_{0}$	2142.3 ± 1.2	2141.4 ^d	3.6 ± 0.1	3.5 ^e
			2141.6^{f}		3.33 ^g
			2141.7 ^h		
			2135.3 ⁱ		
	$2^{3}S_{1}-1^{1}S_{0}$	2126.7 ± 1.2	2124.7 ^d		
		_	2124.8^{f}		
			2124.7 ^h		
			2120.5^{i}		

^aReference 32.

^bReference 34, interpolated value.

^cReference 33, interpolated value; accuracy 0.5 eV.

^dReference 5.

eReference 35.

^fReference 3.

gReference 2.

^hReference 1.

ⁱReference 36, nonrelativistic value.

broadening due to the velocity and angular dispersion of the beam; (ii) an estimated systematic error of $\pm 0.25^{\circ}$ ($\equiv 0.6$ eV) in the beam-detector alignment.

B. Lifetime measurements

Using the high dispersion properties of the spectrometer, it is easy to select a particular x-ray emitter, avoiding multiexponentional fitting procedures. The difference of the x-ray yield on either side of the absorption edge characterizes a particular atomic transition. The lifetime, in the form of a single exponential, is obtained by measuring this difference as a function of the foil-detector separation. The lifetime of the 2^3P_2 state has been measured over a distance of 22.5 cm (corresponding the three mean lines) in steps of 9 mm. The $2^4P_{5/2}$ state was observed over 8 cm (three mean lines) in steps of 4.5 mm. Much care has been taken in the geometrical definition of the beam. The collimators upstream of the spectrometer are experimentally adjusted in such a way that the beam intensity downstream of the spectrometer (FC 2) be independent of the carbon-foil position inside the chamber. Figure 7 is a plot of the brute difference as a function of the foil-detector separation. The straight lines are the best fits obtained with single exponentials. The

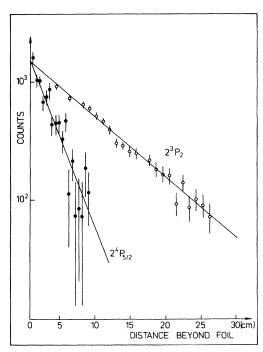


FIG. 7. Decay curves taken with the Doppler-tuned x-ray spectrometer.

experimental lifetimes are

$$\tau(2^3P_2 = (3.6 \pm 0.1) \times 10^{-9} \text{ s}$$

and

$$\tau(2^4P_{5/2}) = (1.2 \pm 0.1) \times 10^{-9} \text{ s}$$
.

The errors are the usual one standard deviations as obtained from the fit.

IV. DISCUSSION

Experimental and theoretical values are confronted in Table I. The ${}^4P_{5/2}$ - ${}^2S_{1/2}$ transition energies are in very good agreement with the theoretical values. The lifetime of the $2{}^4P_{5/2}$ level in Li-like argon has been carefully studied by Dohmann and Mann, 37 taking into account cascading processes. Their uncorrected value is somewhat higher than the theoretical value, 32,34 but when cascading is considered, better agreement is obtained. In our measurement there is no evidence for cascading effects at long distances and the experimental value of the lifetime of the $2{}^4P_{5/2}$ state in P^{12+} is in agreement, within one standard deviation, with the calculations of Ref. 34.

The transition energies in He-like phosphorus are in very good agreement with theoretical predictions. It is shown that the sensitivity of the measurements is such that relativistic effects are easily detected. Finally, the measured lifetime of the 2^3P_2 state is in better agreement with the calculations of Tunell et al.³⁵ than with those performed by Lin and Johnson.²

V. CONCLUSIONS

We have performed energy and lifetime measurements in He-like and Li-like phosphorus. The results are in good agreement with recent theoretical calculations. We have shown that high-resolution x-ray measurements can be achieved by means of a Doppler-tuned x-ray spectrometer; absorption edges in the keV region generally have a natural linewidth in the eV region and angular dispersions down to 1 eV/deg can be easily achieved. We point out, however, that in order to limit systematical effects a critical survey of the tabulated x-ray absorption edges is necessary in light of recent experiments.

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- [‡]National Fund for Scientific Research, Belgium.
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