Transition rates of $1s2s2p$ $^4P_J^o$ states of Li-like ions ($Z = 8,7,6$)

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The radiative and autoionization decay channels of oxygen metastable ionic states have been investigated with beam-foil spectroscopy. Relative branching ratios of the $1s2s2p$ $^4P_{1/2,3/2}^o$ states of O^{5+} to the $J = 5/2$ level have been determined. Radiative and autoionization rates of the $J = 1/2$ and $3/2$ fine-structure levels are extracted by using the calculated fluorescence yield of the $J = 5/2$ level of Chen, Crasemann, and Mark [Phys. Rev. A **27**, 544 (1983)] and the measured total lifetimes corrected for cascades. The study of the delayed electron emission is extended in nitrogen and carbon ions, where transition rates of the dominating autoionization channel of the 2 $^4P_{1/2,5/2}^o$ states have been determined directly from the experimental lifetimes corrected for cascades.

I. INTRODUCTION

The decay of metastable states in few electron ions has long been an interesting subject for both experimental and theoretical studies. In recent years there have been big advances in the theoretical models for the calculation of radiative and autoionization rates of core-excited states of simple configurations, such as the $1snln'l'$ ^4L_J states, in three-electron ions for a wide range of Z. New relativistic calculations in the intermediate-coupling scheme [multiconfiguration Dirac-Fock optimal level[1,2] (MCDF-OL), Dirac-Hartree-Fock-Slater[3] (DHFS), multiconfiguration Dirac-Fock extended average level[4] (MCDF-EAL)] and nonrelativistic saddle-point complex rotation (SPCR) calculation in the intermediate-LSJ-coupling scheme[5] for the three-electron ions give rather different results than previous realistic [$1/Z$ expansion[6]] and nonrelativistic calculations in the LS [Hartree-Slater[7] (HS)] or intermediate coupling [Hartree-Fock-Slater[8] (HFS)] schemes. Discrepancies between the results of most recent calculations are state dependent. Since no fully relativistic configuration-interaction (CI) calculations are routinely available, the electron-electron correlations in the multiconfiguration calculations need further investigation as has been pointed out previously.[9,10] The need for experimental data has been also expressed in several theoretical publications. From the experimental point of view, besides transition energy measurements, the determination of total lifetimes is the most convenient test of the theory.[9] However, if the excited state has more than one decay channel, lifetimes include more than one parameter, i.e., transition rates. Either radiative or autoionization rates can then be directly extracted from the measured lifetimes only if one of the channels dominates overwhelmingly. Furthermore, lifetimes can be strongly affected by cascades and therefore need correction, which can only be done if enough information about the cascades is available. Fluorescence yield measurements have been proposed[11,12] as a possible, cascade-independent, sensitive test of transition-rate calculations.

In a recent publication[12] we investigated fluorescence yields of the $J = 1/2$, $3/2$, and $5/2$ fine-structure components of the $1s2s2p$ $^4P_J^o$ state of Ne^{7+}. X-ray and Auger transition rates of these states have been determined from the fluorescence yields, with the help of the corresponding measured lifetimes corrected for cascades. In the present work we extend our beam-foil studies of the same metastable states for $Z = 8,7,6$ looking into both x-ray and Auger decay channels of O^{5+} and the autoionizing channel of N^{4+} and C^{3+}.

II. EXPERIMENTAL PROCEDURE AND DATA ANALYSIS

A detailed description of the experimental setup and procedure is given elsewhere.[12] The apparatus consists of a plane-crystal x-ray spectrometer [potassium acid phthalate (KAP) crystal] and 30° parallel-plate electron energy analyzer, both equipped with position sensitive detectors and observing the x-ray and Auger electron emission of the same ion beam segment. The excitation of the ions occurred at the carbon foil of a beam-foil apparatus, which permitted delay distances up to 40 cm. The O^+, N^+, and C^+ ion beams were obtained from the 7.5-MV Van de Graaff accelerator of the University of Freiburg at several energies ranging from 1.8 to 5.0 MeV. High-resolution x-ray spectra could be taken only for the oxygen metastable states, since for the other two ions, the photon energies exceed the analyzing range of the crystal spectrometer, determined by its dispersive element and the absorption of the Mylar window of its detector. Hence, only the delayed electron emission of the nitrogen and carbon ions has been observed.

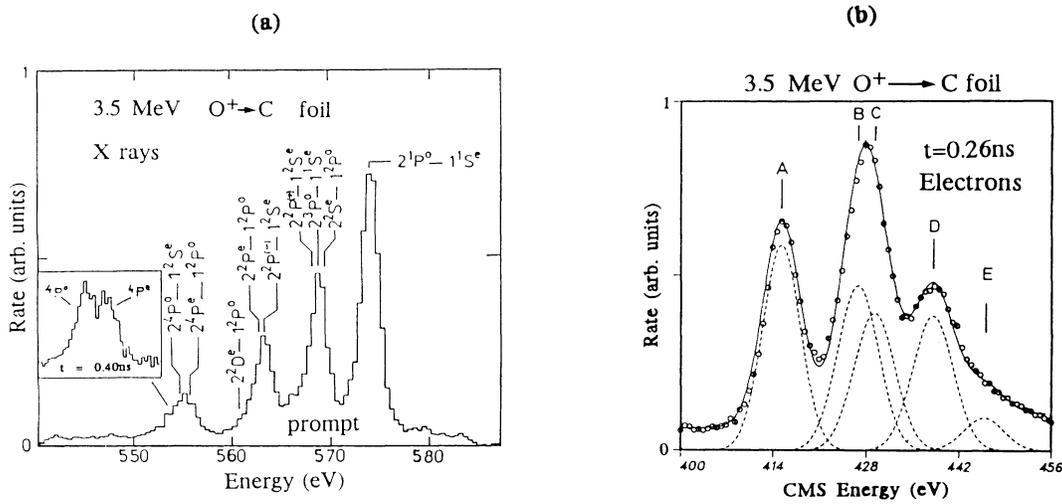Figure 1(a) shows a prompt x-ray spectrum of oxygen

FIG. 1. (a) Oxygen x-ray spectrum in the energy region 540–585 eV. The inserted portion shows the two resolved transitions of the $2\,^4P^o$ and $2\,^4P^e$ states at increased resolution of the crystal spectrometer. (b) Delayed oxygen electron spectrum. The five titled peaks correspond to the transitions A: $2\,^4P^o{-}1\,^2S^e{+}e^-$; B: $2\,^5P^e{-}^2P^o{+}e^-$; C: $2\,^4P^e{-}^1S^e{+}e^-$; D: $2\,^4P^e{-}^2S^e{+}e^-$; E: $2\,^5PS^e{-}^2P^o{+}e^-$.

in the energy region 540–585 eV, which corresponds to the window length of the position sensitive detector. Three peaks remain in the delayed spectra at about 555, 569, and 574 eV, corresponding to the $2\,^4P^o{-}1\,^2S^e$, $2\,^4P^e{-}1\,^2P^o$ (unresolved in the first peak), $2\,^3P^o{-}1\,^1S^e$, and $2\,^1P^o{-}1\,^1S^e$ transitions, respectively. The last allowed $E1$ transition can be seen downstream of the beam, due to the population of the $2\,^1P^o$ levels via cascades.[13] The inserted portion in Fig. 1(a) shows a delayed spectrum at high resolution (1 eV), where the two transitions from the decay of the $2\,^4P^o$ and $2\,^4P^e$ states could be resolved for the first time.

A delayed electron spectrum of oxygen is shown in Fig. 1(b) together with the fit of five peaks A,B,C,D,E and an exponential background. The peaks correspond to the autoionization transitions $1s2s2p\,^4P^o{-}1s^2\,^1S^e{+}e^-$, $1s2s2p^2\,^5P^e{-}1s^22p\,^2P^o{+}e^-$, $1s2p^2\,^4P^e{-}1s^2\,^1S^e{+}e^-$, $1s2s2p^2\,^5P^e{-}1s^22s\,^2S^e{+}e^-$, and $1s2p^3\,^5S^e{-}1s^22p\,^2P^o$ $+e^-$ at about 416, 426, 429, 438, and 445 eV.[14,15] The contribution of sextets is neglected here because the small production of charge state 5 at this ion energy[16] is distributed among a large number of states, each of which undergoes several transitions. Electron spectra of nitrogen and carbon taken at several delay times are shown in Figs. 2(a) and 2(b). The three observed peaks in the nitrogen spectra are due to the same transitions as in the case of oxygen. The absence of the third peak together with the constant position and asymmetry of the right peak in the carbon spectra indicates that, as expected from the low population of charge state 4 at 3 MeV,[16] there is no appreciable intensity contribution from quintet states in the spectra. In contrast, besides the third peak, a shifting and asymmetry change of the central peak is observed for different delay times in the case of nitrogen, oxygen, and neon,[12] due to the different life-

times of the $2\,^4P_j^e$ and $2\,^5P_j^e$ states. For the energy scales in Figs. 2(a) and 2(b) calculated transitions energies[1,2,17] have been used.

The examples of x-ray and electron decay curves of the $^4P_j^o$ states shown in Figs. 3(a) and 3(b) and Figs. 4(a) and 4(b) are obtained from least-square fits to the measured high-resolution spectra. Lifetimes of the three spin-orbit levels are averages obtained from multiexponential least-squares fits of the x-ray and electron decay curves, from several runs at different ion velocities. They are assigned to the $J=1/2$, 3/2, and 5/2 levels after comparison with their most recent theoretical values. They are further corrected for the $E1$ cascade transitions $1s2p^2\,^4P_j^e$ $-1s2s2p\,^4P_j^o$. These are found, as discussed elsewhere,[12] to be the only cascades which can have reasonable influence on the lifetimes of the $2\,^4P_j^o$ states. The correction procedure is based on the comparison of measured with simulated decay curves and is described in a previous publication.[12] Since the branching ratio of the $E1$ decay of $2\,^4P_j^e$ rapidly decreases with Z (Ref. 2) the lifetime corrections become more important for the low Z's, and reach 55% of the measured value for the $J=1/2$ component of carbon. Unlike the case of neon[12] the $E1$ cascade is found to also affect slightly the lifetimes of the $J=3/2$ states of all three ions.

Ratios of the measured x-ray to electron intensities for all J components of oxygen are calculated from the products of the fitted amplitudes (rate at $t=0$, average values) and the corrected lifetimes. The decay of the $2\,^4P_{5/2}$ state can be treated rather accurately theoretically, as it consists only of pure magnetic quadrupole x-ray emission and spin-spin induced magnetic autoionization. Configuration mixing with doublet states, which can make the calculation less reliable, does not contribute to

this decay, as its does for the other two J components. The accurate theoretical treatment of this state can be verified by (a) the similar results for several ions, of a variety of different transition-rate calculations,[1,3,7,8,18] (b) the good agreement of experimental and theoretical lifetimes for a wide range of Z (Refs. 12, 19, 20, and this work) and (c) the fairly good agreement of theoretical ra-

diative and autoionization rates with earlier experimental results in neon,[12] in an experiment at calibrated relative x-ray and electron detection efficiencies. Therefore using the most recent theoretical radiative to autoionization branching ratio of the $J = 5/2$ level, 0.006 75,[1] we obtain branching ratios $R = \Gamma_x/\Gamma_A$ or fluorescence yields $\omega = R/(1+R) = \Gamma_x/\Gamma_x + \Gamma_A$ of the other two fine-

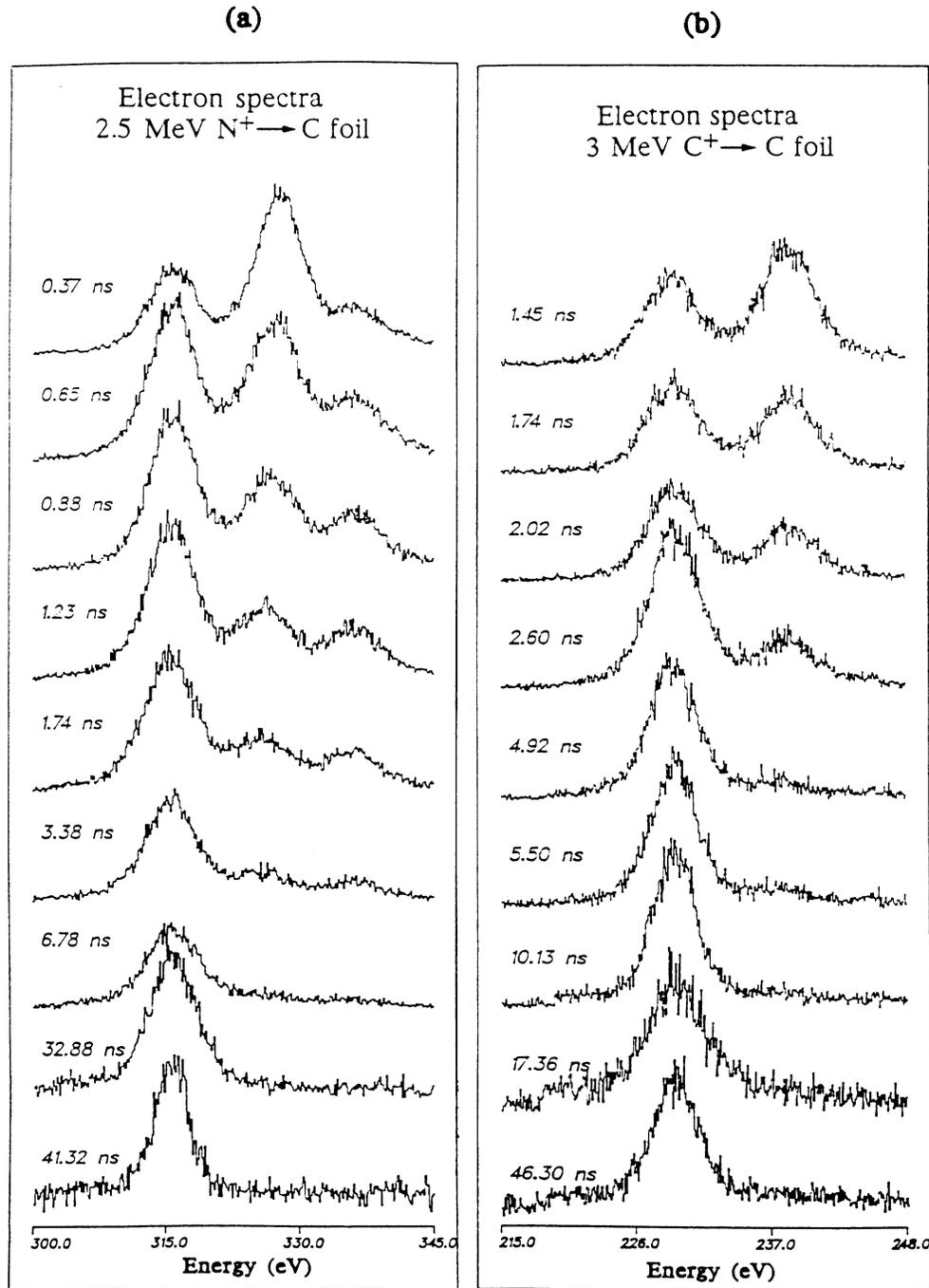**(a)**   **(b)**

FIG. 2. Autoionization electron spectra from foil excited (a) 2.5-MeV N^+ and (b) 3-MeV C^+ beams observed at various delay times downstream of the beam,
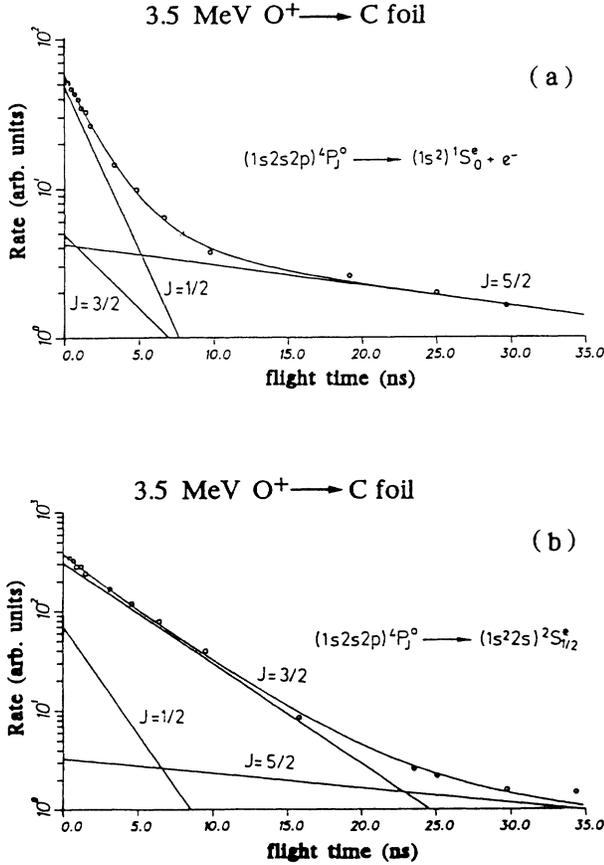
FIG. 3. Autoionization (a) and x-ray (b) decay curves of the oxygen $2\,^4P_J^o$ states and J components obtained from multiexponential fits. Decay rates are given in relative scale.
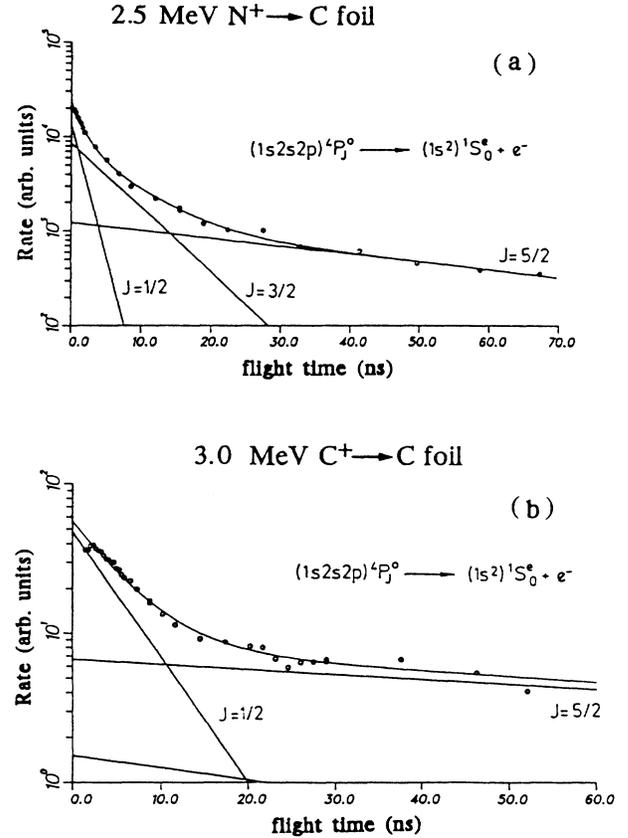
FIG. 4. Autoionization decay curves of the $2\,^4P_J^o$ states of Li-like nitrogen (a) and carbon (b). Decay rates are given in relative scale.

structure levels. Radiative and autoionization rates can then be extracted from the corrected lifetimes by $\Gamma_x = \omega/\tau$, $\Gamma_A = (1-\omega)/\tau$. We have avoided using the relative detection efficiency calibration of the neon experiment[12] because of the large uncertainties which are introduced by the needed correction, when it is applied to the O^{5+} x-ray and Auger energy regions.

For nitrogen and carbon, Auger rates can be derived directly from the measured lifetimes for the $J = 5/2$ component, since for these ions the fluorescence yields are in the order of 0.1%. The same applies with an error $\leq 1\%$ for the $J = 1/2$ states, as it turns out from an extrapola-

tion of the measured radiative and autoionizing rates of this level in oxygen (see below) and neon.[12]

III. RESULTS AND DISCUSSION

A summary of the measured lifetimes, fluorescence yields, radiative and autoionization rates is shown in Table I. The Z dependence of the measured lifetimes and transition rates, for $Z = 10$ (Ref. 12), 8, 7, 6 is shown in Fig. 5(a)–5(c) together with earlier experimental values[11,13,21–23] and results of theoretical calculations.[1,4,6,8]

TABLE I. Measured lifetimes, fluorescence yields, and autoionization and radiative rates of $1s2s2p\,^4P_J^o$ states for $Z = 8,7,6$. Numbers in brackets are powers of 10, e.g., $2.4 \pm 0.7[-8] = (2.4 \pm 0.7) \times 10^{-8}$. 1 a.u. $= 4.134 \times 10^{16}$ s^{-1}.

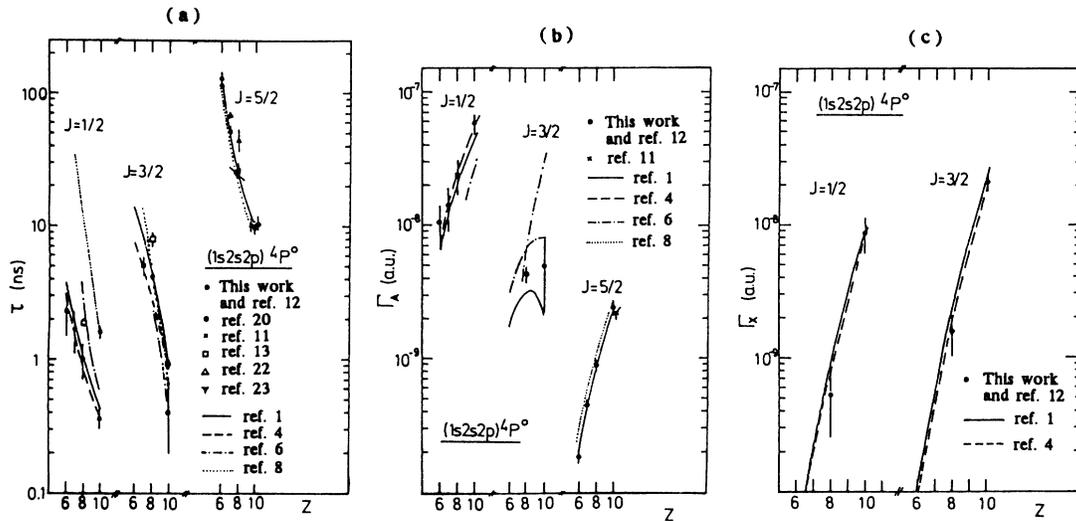|	Oxygen				Nitrogen		Carbon	
J	τ (ns)	ω	Γ_A (a.u.)	Γ_x (a.u.)	τ (ns)	Γ_A (a.u.)	τ (ns)	Γ_A (a.u.)
1/2	1.0 ± 0.3	0.022 ± 0.009	$2.4 \pm 0.7[-8]$	$5.3 \pm 2.7[-10]$	1.7 ± 0.6	$1.4 \pm 0.5[-8]$	2.3 ± 0.8	$1.1 \pm 0.4[-8]$
3/2	4.2 ± 0.3	0.26 ± 0.08	$4.3 \pm 0.6[-9]$	1.5 ± 0.9	5.1 ± 0.9			
5/2	27 ± 3		$9 \pm 1[-10]$		52 ± 5	$4.6 \pm 0.5[-10]$	129 ± 15	$1.9 \pm 0.2[-10]$

FIG. 5. Lifetimes (a), autoionization (b), and radiative rates (c) of fine-structure levels of the $2\,^4P^o$ term for $Z = 10,8,7,6$. Theoretical results are indicated by solid lines (——, MCDF-OL; — — —, MCDF-EAL; —·—·—·, $1/Z$ expansion; · · · ·, HFS). Dots indicate the experimental results of this work including our neon data from Ref. 12. Other experimental data: $\bigcirc$, Schumann et al. (Ref. 20); $\times$, Kricke et al, (Ref. 11); $\square$, Richard et al. (Ref. 13); $\triangle$, Dmitriev et al. (Ref. 22), Pegg et al. (Ref. 23). 1 a.u.$=4.134\times10^{16}$ s^{-1}.

The very good agreement of the measured $J = 5/2$ lifetimes with the theory was expected, since, as discussed above, the theoretical treatment of this state can be more accurate. Also its experimental study is easier than that of the other two J components, as its lifetime is about one order of magnitude longer. Our experimental results are closer to that of the MCDF calculation[1] than to the previous nonrelativistic HS one.[8,1] They are also in good agreement with the experimental values of Pegg et al.[23] for $Z = 10,8$ and agree better with theory for $Z = 8,7$ than the much earlier experiment of Dmitriev et al.[22]

The study of the $J = 1/2$ and $3/2$ transitions is more complicated from both experimental and theoretical points of view. Their similar lifetimes result in large errors in the multiexponential fits. Since configuration mixing with doublet states strongly affects the decay of these states, the results of the transition-rate calculations are very sensitive to the small required doublet to quartet mixing ratios. Hence it is not surprising that, for these mixed states, different calculations give results which differ by up to an order of magnitude (Fig. 5). However, the agreement between the MCDF-EAL (Ref. 4) calculation and our experimental results for the $J = 1/2$ state is fairly good for all four investigated Z's, although, because of the large experimental errors, a strict test of the MCDF-OL (Ref. 1) and MCDF-EAL (Ref. 4) calculations is not possible.

A discrepancy between our results and the calculations of Chen et al.[1,4] is found in the case of the Auger transition rates of the $J = 3/2$ level. For both neon and oxygen our measured values are between the strongly differing results of the OL and EAL calculations.

The large discrepancies between previous experimental values and our $J = 1/2$ and $3/2$ lifetimes for $Z = 10,8$ are due to the interchanged assignment of the $J = 1/2$ and $3/2$ components in the work of Schumann et al.[20] and to

the lower resolution of the x-ray spectrometer in the earlier experiment of Richard et al.[13] In that experiment the $^4P^o_j$ decay curve includes also the three spin-orbit components of the unresolved $2\,^4P^e_{1/2,3/2,5/2}$ decay.

In conclusion, it is obvious that there is better agreement of the MCDF calculations with our experimental results, than those of the earlier nonrelativistic or relativistic $1/Z$-expansion theoretical approaches. By the comparison of our experimental lifetime, fluorescence yield, and transition-rate data with the results of the two OL and EAL calculations, a slightly better overall agreement with the MCDF-EAL calculation is found for the $2\,^4P^o_j$ states of the four investigated Li-like ions. This could be expected as this theoretical approach includes a large number of interacting configurations resulting from the variety of orbitals it uses. However, the accuracy of the calculated atomic state function (ASF) is not obvious since (a) the only criterion for the determination of the orbitals is the minimalization of the average energy of all the states and (b) the exclusion of other configurations than the ones used is not self-evident. As it has been pointed out in a previous publication,[12] we expect that fully relativistic CI calculations would be a more accurate theoretical approach to the problem.

Note added in proof. The most recent SPCR calculations of Davis and Chung[24] are in excellent agreement with our experimental data.

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