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Publication date
1994

Published in
Journal of Physics. B, Atomic, Molecular and Optical Physics

[Link to publication](#)

Citation for published version (APA):

van der Hart, H. W., Vaeck, N., & Hansen, J. E. (1994). Radiative stabilization in double-electron capture to 4l4l' and 4l5l' states in O6+. *Journal of Physics. B, Atomic, Molecular and Optical Physics*, 27(B), 3489-3514.

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Radiative stabilization in double-electron capture to $4\ell 4\ell'$ and $4\ell 5\ell'$ states in O^{6+}

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Received 4 January 1994, in final form 19 April 1994

Abstract. Following slow collisions between O^{8+} and Ar, double-electron capture into $4\ell 4\ell'$ and $4\ell 5\ell'$ states of O^{6+} has been reported. The experimental results show that in 25% of the events the two electrons are retained on the oxygen presumably due to radiative stabilization. In order to understand this high rate of stabilization, we have calculated for the first time the decay properties of all the $4\ell 4\ell'$ and $4\ell 5\ell'$ terms in the isolated atom using a truncated diagonalization method with B -spline basis sets. The $4\ell 4\ell'$ states are lying in the same range of energy as the higher members of the $3\ell n\ell'$ Rydberg series and we have extended our B -spline approach to represent accurately the $3\ell n\ell'$ states with n up to 50. In this static approach, we obtain 30.8% radiative stabilization for the $4\ell 4\ell'$ manifold and 2.6% for the $4\ell 5\ell'$ manifold assuming in both cases a statistical population of the states. We conclude that these values, when combined with the experimentally determined initial populations of the two configurations, are too small to explain the experimental features assuming a statistical population distribution and we propose that in order to clarify the situation an experimental and/or theoretical study of the population distribution over the two configurations is necessary.

1. Introduction

Multiple-charge transfer has been observed in slow collisions between highly charged ions and neutral atoms or molecules. The most probable and most studied of these processes is double capture, DC, of two electrons from the target into excited states of the projectile. According to the extended classical over barrier model, ECBM (Bárány *et al* 1985, Niehaus 1986), which has been very successful in describing the overall features of the process, the capture populates two-electron states $n\ell n'\ell'$ with $n \approx n'$. With increasing projectile charge, the $n\ell n'\ell'$ states become doubly excited and theoretical investigations (Hansen 1989, Luc-Koenig and Bauche 1990) of the decay properties of such states indicate that in a single-configuration approach with a statistical population distribution they decay by autoionization, leading to autoionizing DC (ADC), a conclusion which seemed to be in reasonable agreement with early observations (Groh *et al* 1983, Astner *et al* 1984, Justiniano *et al* 1984, Guillemot *et al* 1990). For example, Astner *et al* (1984) studied the process $Ar^{q+} + Ar \rightarrow Ar^{(q-r)+} + Ar^{2+} + (2-r)e^-$ for q between 2 and 8 and found that the cross section for the $r = 2$ process (T(rue)DC) relative to ADC ($r = 1$) is equal to 19 for $q = 2$ but decreasing to 1.2 for $q = 5$ and further to 0.13 when q increases to 8 reflecting the expectation that the two electrons are picked up in higher and higher and thus autoionizing

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states. The general belief was that this trend would continue with increasing q and in fact Danared *et al* (1987) who studied the same process until $q = 13$ did not report a cross section for the $r = 2$ process for $q \geq 8$. However, recent evidence suggests that at least in some cases this trend can be broken and that certain ions can show much larger degrees of TDC (Stolterfoht *et al* 1990, Roncin *et al* 1991, Cederquist *et al* 1992, Martin *et al* 1992, Morgenstern 1993, Gaboriaud *et al* 1993). In the Ar case, mentioned above, Ali *et al* (1993) have shown that a minimum is reached at $q \approx 9$ and that there is a slight increase thereafter. It is generally assumed that the cause of the 'stabilization' of both electrons on the ion is radiative decay being more probable than autoionization, although evidence for this hypothesis is still primarily circumstantial. (Photons have been observed in the work of Martin *et al* (1992) but an unambiguous connection with TDC was not established.)

Roncin *et al* (1991) noticed that TDC occurs when the states believed to be relevant are very close to or straddling a threshold. For instance, in double-capture processes between O^{8+} and Ar, where Roncin *et al* (1991) and Gaboriaud *et al* (1993) have found that TDC amounts to 25% of the total DC probability, the two electrons are believed to be captured initially in the $4\ell 4\ell'$ or $4\ell 5\ell'$ manifolds. Some $4\ell 4\ell'$ states in O^{6+} lie either just below or just above the $N = 3$ threshold. Therefore, the overlap between the $3\ell n\ell'$ Rydberg series and the $4\ell 4\ell'$ states was proposed by Roncin *et al* (1991) as the likely explanation for the high rate of TDC.

Based on this observation, Bachau *et al* (1992) proposed a mechanism (auto transfer to Rydberg states, ATR), related to the post collision interaction (PCI) effect, in which those $4\ell n\ell'$ states that lie above the $N = 3$ limits in the unperturbed oxygen ion, but below the $N = 3$ limits during the collision, interact with the $3\ell n\ell'$ states during the traverse of the Rydberg series when the projectile is on the way out of the collision. As a result, they can lose their $4\ell n\ell'$ character by mixing with the $3\ell n\ell'$ states that were assumed to stabilize radiatively. To explain the 25% TDC observed experimentally, Bachau *et al* (1992) conjectured that this percentage can be obtained by initial populations of 75% and 25% in the $4\ell 5\ell'$ and $4\ell 4\ell'$ states, respectively, coupled with branching ratios for transfer to Rydberg states of 10% for $4\ell 5\ell'$ and 50% for $4\ell 4\ell'$.

We have commented on this mechanism (Vaeck and Hansen 1993, Vaeck *et al* 1993a, b). We argued that the $4\ell 5\ell'$ states are lying too high above the $N = 3$ threshold to be able to cross this limit even in a molecular picture of the collision process. On the other hand, only five of the 44 terms in the $4\ell 4\ell'$ manifold are located above the $N = 3$ limits in the unperturbed O^{6+} ion and, although we realized that similar effects can be expected for terms lying below but sufficiently close to threshold, we pointed out that in O^{6+} only a very restricted number of states are located in this range of energy. Consequently an additional argument is necessary to explain why these terms would be preferentially populated during the collision. We also showed that the mixing between the $4\ell 4\ell'$ and $3\ell n\ell'$ states is no guarantee for TDC and that it is necessary to consider the properties of individual terms which means that it is difficult to make predictions that are valid for a configuration as a whole. Similarly, also in the absence of dynamic effects, such as the ATR, we noted that certain terms have large radiative branching ratios and the same observation has been made by Chen and Lin (1993). This also means that for this alternative effect it is difficult to make specific predictions unless the population distribution is known.

Thus at the moment two types of radiative stabilization are being considered as possible explanations for the large TDC probabilities observed; indirect stabilization which is a dynamic process, exemplified by the ATR, in which the details of the collision are determining the degree of stabilization and direct stabilization, which is a static process in the sense that it occurs for a particular state in the isolated atom independently of the details of the

collision except insofar as the initial population of the state is due to the collision. In the following, we will call the relevant quantity for the latter process, the radiative branching ratio $R^r = A^r/(A^a + A^r)$ the 'stabilization ratio'. Here A^r is the radiative decay rate and A^a the autoionization rate. We will also use the same term for the indirect stabilization process and, in general, we will use the expression 'stabilization' as a synonym for TDC.

In their recent experimental work Gaboriaud *et al* (1993) found considerable probabilities for TDC for the collision systems $N^{7+} + \text{Kr}$ (at 10.5 keV), $O^{8+} + \text{Ar}$ (at 12 keV) and $Al^{9+} + \text{Ne}$ (at 9 keV). Gaboriaud *et al* reported evidence for direct stabilization of the $4\ell 5\ell'$ states while interpreting the stabilization for the $4\ell 4\ell'$ manifold as being due to indirect stabilization. More quantitative analyses of indirect stabilization for the $4\ell 4\ell'$ states populated during the $N^{7+} + \text{He}$ and Ar collision processes experimentally, as well as theoretically, were carried out by Bachau *et al* (1993) and Roncin *et al* (1993) who emphasized the importance for the ATR process of the states immediately below threshold in addition to those above. Taking this into account, they obtained 18% radiative stabilization for the singlet $4\ell 4\ell'$ terms in N^{5+} . In N^{5+} the $4\ell 4\ell'$ terms are located higher compared to the $N = 3$ threshold than in O^{6+} so that more terms are located in the region favourable to the ATR effect, but although 18% is an appreciable amount, it is far from the >52% deduced from experiment for both singlet and triplet terms in N^{5+} (Gaboriaud *et al* 1993). It is clear that there is an uncertainty in the theoretical value due to the complexity of the calculations but, since we already have pointed out that direct stabilization can be increased at least for some of the $4\ell 4\ell'$ states below the $N = 3$ threshold due to mixing with the $3\ell n\ell'$ states, we have found it of interest to try to quantify this alternative effect. Surprisingly, we find a larger stabilization ratio for O^{6+} in this purely static approach.

The results, which are reported here, represent the first complete study of the $4\ell 4\ell'$ and $4\ell 5\ell'$ manifolds in the free O^{6+} ion. Using an extension of our *B*-spline technique (Hansen *et al* 1993) to take into account the mixing with the $3\ell n\ell'$ Rydberg series for very high n values, we have investigated the decay properties of these states as well as the reliability of the scaling law for the $3\ell n\ell'$ Rydberg states. We also compare our results with predictions due to Chen and Lin (1993) made on the basis of the $(K, T)^A$ scheme. Our results confirm our previous observations concerning the stabilization process in double-electron capture (Vaeck and Hansen 1993, Vaeck *et al* 1993a, b) and show large stabilization ratios for particular terms in these configurations (in particular the singlet terms with parity $\Pi = (-1)^{L+1}$) even for the $4\ell 5\ell'$ states which are not degenerate with the $3\ell n\ell'$ Rydberg series. The numbers presented here supersede the preliminary values in Vaeck *et al* (1993a, b).

At the moment very little is known about the actual population distribution in the pickup and in the calculations in this paper we will assume that the distribution over a particular configuration is statistical. The validity of this assumption will be discussed briefly in chapter 4. In the following we also present averages over various subsets of the terms within a particular $n\ell n'\ell'$ manifold using in each case a statistical population distribution and we will call the average stabilization ratio for the whole manifold in this approximation the 'overall' ratio.

2. Calculational approach

2.1. Truncated diagonalization method

To determine the properties of the $4\ell 4\ell'$ and $4\ell 5\ell'$ states in O^{6+} , we have used an approach based on *B*-spline basis functions to describe the different types of wavefunctions involved. *B*-splines allow us to describe doubly-excited as well as Rydberg and continuum states

(Hansen *et al* 1993) in such a way that it is easy to determine their mutual interaction. Using the truncated diagonalization method (TDM) discussed earlier (van der Hart and Hansen 1992a), we have determined energies and wavefunctions of the $4\ell 4\ell'$ and $4\ell 5\ell'$ states. In this approach the interaction with the $(1, 2, 3)\ell(n/\epsilon)\ell'$ series is neglected. The mono-electronic wavefunctions are hydrogenic functions which implies that we do not have to include overlap matrix elements in the calculations, the orthogonality of the two-electron wavefunctions being ensured automatically. The size of the matrices involved is between 1300 and 2500. Since we are only interested in the low-lying states, we have used a modified Davidson routine (Davidson 1975) to determine only the lowest eigenvectors and eigenvalues. The *B*-splines used in this part of the calculation are of order 7. They are defined on an interval from 0 to 40 au divided into 21 segments using an exponential knot point distribution. The configuration expansions used in these calculations are listed in table 1.

Table 1. Basis sets used in the expansion of the $4\ell(4, 5)\ell'$ wavefunctions. Each basis set consist of 25 splines but the number shows the highest spline solution which is used in the calculation for the particular *LS* term.

<i>LS</i>	Configurations	s	p	d	f	g	h	i	k	l
$1S^e$	$s^2, p^2, d^2, f^2, g^2, h^2, i^2$	25	25	25	25	25	25	25		
$3S^e$	$s^2, p^2, d^2, f^2, g^2, h^2, i^2$ ^a	25	25	25	25	25	25	25		
$1P^o$	sp, pd, df, fg, gh, hi	22	22	22	22	20	18	16		
$3P^o$	sp, pd, df, fg, gh, hi	22	22	22	22	20	18	16		
$1P^e$	$p^2, d^2, f^2, g^2, h^2, i^2$ ^a	0	20	20	18	16	16	10		
$3P^e$	$p^2, d^2, f^2, g^2, h^2, i^2$	0	20	20	18	16	16	10		
$1D^o$	pd, df, fg, gh, hi	0	20	20	18	16	16	10		
$3D^o$	pd, df, fg, gh, hi	0	20	20	18	16	16	10		
$1D^e$	$sd, p^2, pf, d^2, dg, f^2, fh, g^2, gi, h^2$	20	20	20	18	16	16	10		
$3D^e$	$sd, p^2, pf, d^2, dg, f^2, fh, g^2, gi, h^2$ ^a	20	20	20	18	16	16	10		
$1F^o$	$sf, pd, pg, df, dh, fg, fi, gh, hi$	20	20	20	18	16	16	10		
$3F^o$	$sf, pd, pg, df, dh, fg, fi, gh, hi$	20	20	20	18	16	16	10		
$1F^e$	$pf, d^2, dg, f^2, fh, g^2, gi, h^2$ ^a	0	20	20	18	16	16	10		
$3F^e$	$pf, d^2, dg, f^2, fh, g^2, gi, h^2$	0	20	20	18	16	16	10		
$1G^o$	$pg, df, dh, fg, fi, gh, hi$	0	20	20	18	16	16	10		
$3G^o$	$pg, df, dh, fg, fi, gh, hi$	0	20	20	18	16	16	10		
$1G^e$	$sg, pf, ph, d^2, dg, di, f^2, fh, g^2, gi$	20	20	20	18	16	16	10		
$3G^e$	$sg, pf, ph, d^2, dg, di, f^2, fh, g^2, gi$ ^a	20	20	20	18	16	16	10		
$1H^o$	$sh, pg, pi, df, dh, dk, fg, fi, gh, gk$	20	20	20	18	16	16	10	10	
$3H^o$	$sh, pg, pi, df, dh, dk, fg, fi, gh, gk$	20	20	20	18	16	16	10	10	
$1H^e$	$ph, dg, di, f^2, fh, g^2, gi, h^2$ ^a	0	20	20	18	16	16	10		
$3H^e$	$ph, dg, di, f^2, fh, g^2, gi, h^2$	0	20	20	18	16	16	10		
$1I^o$	$si, ph, pk, dg, di, dl, f^2, fh, fk, g^2, gi, gl$	20	20	20	18	16	14	12	10	10
$3I^o$	$si, ph, pk, dg, di, dl, f^2, fh, fk, g^2, gi, gl$ ^a	20	20	20	18	16	14	12	10	10
$1I^e$	$pi, dh, dk, fg, fi, fl, gh, gk$	0	20	20	18	16	14	12	10	10
$3I^e$	$pi, dh, dk, fg, fi, fl, gh, gk$	0	20	20	18	16	14	12	10	10
$1K^o$	$sk, pi, pl, dh, di, fg, fi, fl, gh, gk$	20	20	20	18	16	14	12	10	10
$3K^o$	$sk, pi, pl, dh, di, fg, fi, fl, gh, gk$	20	20	20	18	16	14	12	10	10

^a In $n\ell n'\ell'$ configurations of this symmetry, configurations with $n = n'$ are not included in the two-electron basis.

Most of the $4\ell 4\ell'$ states in O^{6+} are, as mentioned, degenerate with the $3\ell n\ell'$ Rydberg series. Therefore in order to represent the interaction between the $4\ell 4\ell'$ states and these Rydberg series, accurate wavefunctions for the $3\ell n\ell'$ states are necessary in some cases for

very large n values. The 3ℓ wavefunctions can be obtained from the B -spline set used in the determination of the $4\ell 4\ell'$ states but to obtain energies and wavefunctions for the $n\ell$ series, we have used a B -spline basis set consisting of 199 splines of order 9 defined on the interval from 0 to 500 au with an exponential knot distribution. This enabled us to accurately determine all states up to $n = 40$. An exponential knot set is used because the Rydberg states, in contrast to continuum states, do not oscillate rapidly at large distance. For the $^1G^e$ Rydberg series, it was found necessary to go to higher n values, the highest $4\ell 4\ell'$ $^1G^e$ term lying very close to the $N = 3$ threshold of O^{7+} (Vaecq *et al* 1993a). For this symmetry, 181 B -splines of order 9 are used, defined on an interval from 0 to 1000 au, yielding accurate wavefunctions for states with n up to 50. The Rydberg series for the different LS symmetries are shown in table 2.

Table 2. The $3\ell n\ell'$ Rydberg series corresponding to all LS symmetries for which at least one $4\ell 4\ell'$ state exist. For each symmetry, the number of $4\ell 4\ell'$ states and their average radiative decay rate ($A_{4\ell 4\ell'}^r$ in 10^{11} s^{-1}) are shown together with the number and types of series and the average radiative decay rate (A_{Ryd}^r in 10^{11} s^{-1}) for each series. In the radiative decay rates the interaction between $4\ell 4\ell'$ and $3\ell n\ell'$ is neglected. For the method of calculation see the text.

LS	No. $4\ell 4\ell'$	$A_{4\ell 4\ell'}^r$	No series	Type of Rydberg series	A_{Ryd}^r
$^1S^e$	4	2.45	3	3sns, 3pnp, 3dnd	3.56
$^1P^o$	3	2.99	5	3snp, 3pns, 3pnd, 3dnp, 3dnf	4.22
$^3P^o$	3	2.93	5	3snp, 3pns, 3pnd, 3dnp, 3dnf	4.22
$^3P^e$	3	3.05	2	3pnp, 3dnd	5.21
$^1D^o$	2	2.77	3	3pnd, 3dnp, 3dnf	4.36
$^3D^o$	2	3.02	3	3pnd, 3dnp, 3dnf	4.36
$^1D^e$	5	2.80	6	3snd, 3pnp, 3pnf, 3dns, 3dnd, 3dng	3.96
$^3D^e$	2	2.34	6	3snd, 3pnp, 3pnf, 3dns, 3dnd, 3dng	3.96
$^1F^o$	3	2.12	6	3snf, 3pnd, 3png, 3dnp, 3dnf, 3dnh	3.96
$^3F^o$	3	2.07	6	3snf, 3pnd, 3png, 3dnp, 3dnf, 3dnh	3.96
$^1F^e$	1	3.46	3	3pnf, 3dnd, 3dng	4.36
$^3F^e$	3	2.15	3	3pnf, 3dnd, 3dng	4.36
$^1G^o$	1	1.45	3	3png, 3dnf, 3dnh	4.37
$^3G^o$	1	1.49	3	3png, 3dnf, 3dnh	4.36
$^1G^e$	3	2.15	6	3sng, 3pnf, 3pnh, 3dnd, 3dng, 3dni	3.96
$^3G^e$	1	3.43	6	3sng, 3pnf, 3pnh, 3dnd, 3dng, 3dni	4.00
$^1H^o$	1	1.47	6	3snh, 3png, 3pni, 3dnf, 3dnh, 3dnk	3.96
$^3H^o$	1	1.42	6	3snh, 3png, 3pni, 3dnf, 3dnh, 3dnk	3.96
$^3H^e$	1	0.93	3	3pnh, 3dng, 3dni	4.36
$^1J^o$	1	0.85	6	3sni, 3pnh, 3pnk, 3dng, 3dni, 3dnl	3.96

The interaction between the $3\ell n\ell'$ Rydberg series and the $4\ell 4\ell'$ states is calculated in two steps. First the eigenvector of each $4\ell 4\ell'$ state is obtained from the TDM described above and secondly the interaction between each of these eigenstates and the Rydberg series is determined followed by the final diagonalization with all the $3\ell n\ell'$ series of the same symmetry included but with only the lowest 141 basis states for each series taken into account. For the $^1G^e$ series, all basis states are taken into account. Due to the near coincidence in energy between some of the $4\ell 4\ell'$ terms and the closely spaced $3\ell n\ell'$ series, it is an approximation to freeze the $4\ell 4\ell'$ eigenvector composition in the calculation of the interaction between the $4\ell 4\ell'$ and the $3\ell n\ell'$ states. It is done in order to reduce the size of the interaction matrices since in these calculations all eigenvalues and eigenvectors are needed. The matrix sizes lie between 300 and 850 depending on the number of Rydberg series with the particular symmetry. For the $^1G^e$ symmetry the matrix has a size of 1150.

Although the $4\ell 4\ell'$ and the higher $3\ell n\ell'$ states are located in the same range of energy they are located in different regions of space which justifies the use of different cavity radii for the 4ℓ and $n\ell$ electrons. The $4\ell 4\ell'$ states are easily confined within a cavity of 40 au, so that it is a good approximation to set the 4ℓ wavefunctions outside this cavity to zero. However, to determine the interaction between different $3\ell n\ell'$ states it is necessary to integrate over the entire cavity, i.e. 500 au.

Finally, the energy shifts due to the $(1, 2)\ell(n/\varepsilon)\ell'$ states are determined for all $4\ell 4\ell'$ and $3\ell n\ell'$ terms by a perturbative approach (van der Hart and Hansen 1992b). In these calculations, another set of B -splines is necessary to determine accurate continuum wavefunctions. Since continuum functions oscillate over the entire cavity range we use a composite knot set (van der Hart and Hansen 1992b) which is exponential near the nucleus and linear from 0.6 au to the boundary of the cavity. A total of 141 B -splines of order 7 is generated with a cavity radius of 40 au for continuum electrons coupled to terms with $L = S$ to G and a cavity radius of 80 au for terms with $L = H$ to K . The number of B -splines is the same in both cavities. This basis set allows us to describe continuum energies up to about 35 au which is necessary for the calculation of the autoionization rates described in the next section.

2.2. Autoionization decay rates

The autoionization rates are determined perturbatively via Fermi's golden rule (van der Hart and Hansen 1992b). For the $4\ell 4\ell' + 3\ell n\ell'$ states the decays to the $1s$, 2ℓ and (for a few terms the 3ℓ) limits are taken into account. The free electron energies lie between 0.003 and 30 au. It can be seen from the above that the continuum cavity radius is much smaller than the cavity radius used for the Rydberg states with high values of n . However, knowledge about the continuum wavefunctions beyond the region of space where the lowest bound functions are localized is unnecessary for the calculation of the autoionization rates so that the difference in box size does not affect the calculation.

The results obtained with this method (van der Hart and Hansen 1992b, 1993a, b) have shown that the accuracy of the total autoionization rates is generally of the order of a few per cent if the independent channel approximation, ICA, is valid. It has been shown, that for systems, which are ionized more than a few times, the effect of the coupling between the different open channels on the total widths is small for most states and that only for a fairly small number of states which have both electrons located preferentially on the same side of the nucleus, so-called 'anti-Wannier states', the coupling becomes important (Martín *et al* 1989, 1991). Since in the present context only total widths are important, use of the ICA is justified but if partial widths are involved, for example, if only autoionization towards $2p$ is monitored, the coupling has to be taken into account. Also for those $4\ell 4\ell'$ states which are located just above the $N = 3$ threshold interchannel coupling might be important for the autoionization rates. However, the resulting effect on the stabilization ratios is very small, since the autoionization rates dominate totally for most of these states as mentioned later. For the lowest states of each symmetry, belonging to the $3\ell 3\ell'$ configurations, our total widths are in good agreement with those reported by Bachau *et al* (1991).

The effect of the coupling between a $4\ell 4\ell'$ LS term and a Rydberg series is illustrated in figure 1 for the 1^1F^o term. There are six $3\ell n\ell'$ Rydberg series with the 1^1F^o symmetry but only one of these has a significant interaction with the $4\ell 4\ell'$ term and only this series is shown in figure 1, that (above) shows the amount of $4\ell 4\ell'$ in each eigenstate of the perturbed 1^1F^o series and (below) the corresponding autoionization decay rate. The calculated autoionization rates show, as expected, a Fano-type profile with cancellation (leading to stabilization) on the

Figure 1. The effect of the interaction between a $4\ell 4\ell'$ perturber, the $1I^e$ term, and a Rydberg series on the autoionization decay rates. In reality there are six Rydberg series with $1I^e$ symmetry but only one of these interacts with the $4\ell 4\ell'$ term and for clarity only this series is shown in the figure. The amount of the perturber in the individual states is shown in the upper panel (crosses) and the corresponding autoionization rates, $A \equiv A_{\text{WI}}^0$ in au, in the lower one (squares). Note that the maximum amount of $1I^e$ character in any state is less than 7%. In the Rydberg series, $n = 13$ corresponds to $E \approx -3.7$ au.

low energy side and constructive interference on the high energy side of the resonance. The detailed structure depends on the relative strengths and phases of the matrix elements coupling the perturber and the series to the continuum and to each other as well as on the position of the perturber in the series. The $1I^e$ term is located rather high in the Rydberg series and the interaction is fairly strong which leads to a smoother distribution of autoionization rates than if the perturber is located lower in the series but the same general behaviour is observed. In the chosen example the perturber interacts nearly exclusively with one series but in other cases where the interaction is with several continua the final result might be less transparent (see, for example, figure 5).

For the determination of the autoionization widths of the $4\ell 5\ell'$ states, we have only considered the decay to the $3\ell\epsilon\ell'$ continua, since autoionization to the closer limit is dominant. Also for these states, we are only interested in the total decay rates and we have therefore not included the coupling between the open channels. The $4\ell 5\ell'$ states lie more than 11 eV above the $N = 3$ threshold so that interchannel coupling is expected to have a small effect on the total decay rate except perhaps for a few anti-Wannier states.

2.3. Radiative decay rates and stabilization ratios

Inside a particular manifold, the radiative decay rates for different terms generally differ by less than one order of magnitude, whereas the autoionization rates can differ by several orders of magnitude. Therefore detailed knowledge of the radiative rates is less important than knowledge about the autoionization rates. At the same time a detailed calculation of the radiative decay rate is more difficult than the calculation of the autoionization rate since wavefunctions for a very large number of possible final states are required which necessitates a large number of separate calculations. Nevertheless reasonably accurate values are required and we have tried to make sure that our radiative rates are accurate.

The eigenvectors of the $4\ell 5\ell'$ states are principally dominated by the contributions of configurations belonging to the Layzer complex. We calculated the radiative decay rates of the $4\ell 5\ell'$ states using the suite of programs written by Cowan (1981). In this approach the basis functions are HF functions optimized on the average energy of each configuration and we limited the CI expansion to the configurations included in the Layzer complex. For the $^1S^e$ symmetry we also performed a complete B -spline calculation and compared the radiative decay rates with the CI values. The agreement between the two approaches was very good confirming the validity of the CI results in this case.

For the $4\ell 4\ell'$ states the problem is much more difficult since the $4\ell 4\ell'$ states interact strongly with a large number of $3\ell n\ell'$ states. This gives rise to very complex eigenvectors in which the $4\ell 4\ell'$ character can be small. Moreover very little is known about the radiative decay rates of the $4\ell 4\ell'$ states even when the interaction with the Rydberg series is neglected. Roncin *et al* (1993) estimated the radiative decay rates of the $4\ell 4\ell'$ singlet terms in N^{5+} interacting with the $3\ell n\ell'$ Rydberg series by considering only the decay of a hydrogenic 3ℓ electron in order to emphasize the importance of the ATR mechanism. We have calculated separately the decay of the $4\ell 4\ell'$ states (in a CI calculation including only the configurations belonging to the Layzer complex) and the radiative decay rates of the $3\ell n\ell'$ Rydberg series. We find for O^{6+} , that the radiative decay rates of the $4\ell 4\ell'$ states are comparable with those for the $3\ell n\ell'$ series. For the $4\ell 4\ell'$ terms the slowest radiative decay has been found for the $^1I^e$ term ($8.54 \times 10^{10} \text{ s}^{-1}$) while the largest radiative rate is found for the lowest $^3P^e$ state ($4.75 \times 10^{11} \text{ s}^{-1}$) and 11 other states have rates larger than $3 \times 10^{11} \text{ s}^{-1}$. Also in this case we calculated the radiative decay rates for the $^1S^e$ symmetry (neglecting the interaction with the $3\ell n\ell'$ series) using the B -spline approach and comparison with the CI results shows good agreement. The rates for the individual terms are used in the calculation of the stabilization ratios for the $4\ell 4\ell'$ terms, when the interaction with the $3\ell n\ell'$ series is neglected, while we have used the average rates for each symmetry when the interaction with the series is taken into account as described below. The average rates are given in table 2.

For the Rydberg series the radiative decay rate is totally dominated by the decay of the 3ℓ electron. We estimated the decay rate by considering the decay of the 3ℓ electron towards $(1, 2)\ell$ using a CI expansion including for each LS symmetry all the $3\ell 14\ell'$ states which can interact with the $4\ell 4\ell'$ states. Our previous mono-configurational calculation for the radiative decay of the $3s18s \ ^1S$ state in O^{6+} (Vaeck *et al* 1993a) showed that only the decay of the inner electron is important and the decay rate is in very good agreement with the hydrogenic value. However, a considerable variation is possible between states with the same LS symmetry depending mainly on the relative weights of the $3sn\ell$ and the $3pn\ell$ components, which have the smallest and the largest decay rates, respectively. Nevertheless, the variation in the average radiative decay rate for each LS symmetry is much smaller and is, in fact, in very good agreement with a simple hydrogenic estimate principally because interference between the different decays is limited by the fact that $3p \rightarrow 1s$ dominates. Also these average rates are given in table 2.

The radiative decay rate for each state has been calculated from

$$A^r = A_{\text{Ryd}}^r + c(4\ell 4\ell')^2 [A_{4\ell 4\ell'}^r - A_{\text{Ryd}}^r] \quad (1)$$

where $c(4\ell 4\ell')^2$ is the total weight of the $4\ell 4\ell'$ components in the eigenvector and A_{Ryd}^r and $A_{4\ell 4\ell'}^r$ the average radiative decay rate for the Rydberg series and for the $4\ell 4\ell'$ states of the particular LS symmetry (table 2), respectively. Equation (1) neglects the interference between the different components of the eigenvector but insofar as the $4\ell 4\ell'$ and $3\ell n\ell'$ basis states decay to different final states there is no interference between these

contributions. On the other hand, the use of average rates in (1) is one of the major approximations in the present work and one reason that we do not show R^c values for each $4\ell 4\ell'$ term, but only results averaged over all terms of the same symmetry. The other reason is that, assuming a statistical population distribution, the average value for each symmetry is the physically relevant quantity. We believe that these values are quite accurate, since in calculations for Ne^{8+} (van der Hart *et al* 1994) we have used (1) but with the radiative rates for the individual $4\ell 4\ell'$ terms instead of the average values. The difference between the two approximations was less than 0.5% in the average value for each symmetry.

Using the rates in table 2, we have estimated a fluorescence yield for each LS symmetry in the following way. Because of the dispersion of the $4\ell 4\ell'$ states within the $3\ell n\ell'$ series, we have considered all states of a particular symmetry whether the main component is $4\ell 4\ell'$ or $3\ell n\ell'$. The stabilization ratio for each term is determined and multiplied by the $4\ell 4\ell'$ weight in the particular state. All the contributions for each LS symmetry, whether above or below the $N = 3$ threshold, are summed and divided by the number of $4\ell 4\ell'$ states present in this symmetry. To obtain the overall ratio, an average over all LS symmetries is carried out using a $(2S + 1)(2L + 1)$ weighting factor. Although the stabilization ratios for the individual terms (not shown) probably can have a non-negligible error we believe that the error in the average values is considerably smaller. One reason is that in practise the influence of the $4\ell 4\ell'$ rates on the (average) stabilization ratios is rather small. An indication of the sensitivity of the 'overall' stabilization ratio to the assumptions about the radiative decay rates can be obtained by noting that use of a common radiative decay rate of $3.5 \times 10^{11} \text{ s}^{-1}$ (the average hydrogenic value for a 3ℓ electron) for all states, instead of using (1), leads to a change of only 0.2% in our final overall value.

The final 'overall' result corresponds to the radiative branching ratio of the $4\ell 4\ell'$ manifold in O^{6+} assuming that the capture process populates the $4\ell 4\ell'$ basis states according to their statistical weight, that the $3\ell n\ell'$ basis states are not populated and, finally, that the decay takes place in the free atom.

It is important to note the difference between the radiative decays $3p \rightarrow 1s$ and $3\ell \rightarrow 2\ell'$. The decay of a $3\ell n\ell'$ state to $1sn\ell'$ leads directly to stabilization of the two electrons while if the decay is to a $2\ell n\ell'$ state the situation is less clear. The $2\ell n\ell'$ states are located in the $1s\ell$ continuum and can therefore decay either by autoionization to the $1s$ limit or by radiative decay to a $1sn\ell'$ state. However, in the present context it is fortunate that the radiative decay rate for a transition $2pn\ell \rightarrow 1sn\ell$ is very large compared to the probability for autoionization when the Rydberg electron has a high n or ℓ value. For $2pn\ell$ states in C^{4+} (van der Hart and Hansen 1993a) and in N^{5+} (Vaeck and Hansen 1992) it has been found that the radiative decay rates are larger than the autoionization rates for all terms already for n less than 10 and generally the mixing between $2pn\ell$ and $2sn\ell'$ is so large in a two-electron atom, due to the degeneracy between the hydrogenic $2s$ and $2p$ states, that the same will apply to most $2\ell n\ell'$ states. Similarly, for the $3\ell n\ell'$ states the weight of the $3pn\ell$ component in the eigenvector is often large enough to allow the direct decay to a $1sn\ell$ state. Therefore, we assume that the second step is not important for the calculation of the stabilization ratios in a two-electron atom.

The calculations of the $4\ell 4\ell'$ and the $4\ell 5\ell'$ energies and the autoionization rates of $4\ell 5\ell'$ states were performed on the Cray YMP4/484 in Amsterdam. A typical run requires less than two minutes of CPU time. The determination of the interaction between the $3\ell n\ell'$ Rydberg series and the $4\ell 4\ell'$ states and their autoionization widths were performed on SUN SPARCstations SLC and ELC. These calculations required up to 10 hours.

3. Results

3.1. The $4\ell 4\ell'$ configurations

Using coincident energy gain spectroscopy, Roncin *et al* (1991) and Gaboriaud *et al* (1993) have observed 25% TDC in collisions between bare O^{8+} ions and Ar and deduced that this corresponds to a stabilization ratio of 59% for the $4\ell 4\ell'$ states and 11% for the $4\ell 5\ell'$ states (Gaboriaud *et al* 1993). For $n\ell n'\ell'$ states with $n \approx n'$, this observation is in stark contrast to the results of previous calculations although little specific knowledge about $n = 4$ systems exist. For example, for the singlet $2\ell 2\ell'$ and $3\ell 3\ell'$ terms in C^{4+} radiative stabilization has been found theoretically to amount to only 2% and 0.3%, respectively, assuming a statistical population distribution (van der Hart and Hansen 1993a). For higher $n\ell n'\ell'$ states, autoionization decay rates are expected to increase since the number of limits increases (Hansen 1989) but very few calculations have actually been carried out for intermediate values of n . In particular, very little is known about radiative decay probabilities.

Figure 2. Energy positions of the $4\ell 4\ell'$ terms in O^{6+} without inclusion of the interaction with the $3\ell n\ell'$ series. Broken lines are used to represent terms of the unnatural parity ($\pi = (-1)^{L+1}$) while the natural parity terms ($\pi = (-1)^L$) are represented by full lines. The $N = 3$ threshold and the average $3\ell 18\ell'$ energy are also indicated.

A schematic representation of the energy positions of the 44 $4\ell 4\ell'$ states is given in figure 2. The figure shows the $4\ell 4\ell'$ energies without inclusion of the interactions with the $3\ell n\ell'$ Rydberg series. The correction to the energy due to the $(1, 2)\ell(n, \varepsilon)\ell'$ bound states and continua is very small ($\pm 1 \times 10^{-4}$ au) for all states while the correction due to the interaction with the $3\ell n\ell'$ Rydberg series is much larger and in fact large enough to push the highest ${}^3P^e$ term above the $N = 3$ threshold. Broken lines are used to represent states of the so-called unnatural parity ($\pi = (-1)^{L+1}$) while the natural parity states ($\pi = (-1)^L$) are represented by full lines. The $N = 3$ threshold and the $3\ell 18\ell'$ energies are also indicated, since $n = 18$ was estimated by Bachau *et al* (1992) as the limit beyond which the high Rydberg series could be considered as a low continuum. Figure 2 shows that, as mentioned already, the $4\ell 4\ell'$ states overlap the $3\ell n\ell'$ Rydberg series in the free O^{6+} ion and the effect of this degeneracy on the decay properties of the $4\ell 4\ell'$ states depend on the unperturbed position of the $4\ell 4\ell'$ terms.

Figure 3. Distribution of the $4l4l'$ character over the six $3lnl'$ Rydberg series for the three $^1G^e$ states in O^{6+} . In the Rydberg series, $n = 13$ corresponds to $E \approx -3.7$ au.

The $4l4l'$ states can be divided into three categories: (i) the states above the $N = 3$ threshold (five terms including $^3P^e$), (ii) the states below the $N = 3$ threshold but above the $3l18l'$ states (eight terms), and (iii) the 31 terms lying below the $3l18l'$ Rydberg states.

The five highest states (the highest $^1S^e$, $^1P^o$, $^1D^e$, $^1F^o$ and $^3P^e$ terms) are located above the $N = 3$ threshold and the autoionization rates of these terms are therefore very large in comparison with the radiative rates. Consequently, these terms will not contribute to the radiative stabilization of the $4l4l'$ manifold. In contrast the ATR mechanism was expected to be particularly effective for these terms.

The second type of state is lying in a region of energy where the density of $3lnl'$ states is very high. These states are considered by Bachau *et al* (1993) and Roncin *et al* (1993) to be completely 'smeared into' the Rydberg series giving rise to a kind of resonance-like structure. We have observed this behaviour for nearly all the *natural* parity states lying below the $N = 3$ threshold but above the $3l18l'$ Rydberg states, i.e. the $^1I^e$ (figure 1), the $^1H^o$, the third $^1G^e$ (figure 3) and the third $^1S^e$ terms. For all these states the $4l4l'$ weight in a particular level is less than 10%. The third $^1G^e$ term is located very close to the limit and is the most difficult to describe. In the present calculation, it has been necessary to extend the radius of the cavity considerably to obtain a good description of this term, which is smeared out over a large number of states with a maximum $4l4l'$ weight of less than 3% (figure 3). For the fourth $^1D^e$ and the third $^3P^o$ term, although the same type of behaviour is observed, the largest $4l4l'$ weight in a particular state is about 20%. On the other hand, the *unnatural* parity states of this type, the second $^3D^o$ and the third $^3F^e$ terms, have a surprisingly large $4l4l'$ percentage, 70% and 40%, respectively. For the $3lnl'$ singlet terms in C^{4+} , van der Hart and Hansen (1993a) have observed that the interaction with the $2l\epsilon l'$ continua is weaker for the unnatural parity states than for the natural parity states leading to larger fluorescence yields for the former.

It is not particularly meaningful to choose a particular state as the perturbed $4l4l'$ term, when no state has more than 25% $4l4l'$ character, say. Only properties averaged over all states are relevant (table 3). However, we note that the results for the individual terms show

Table 3. Stabilization ratios for the $4\ell 4\ell'$ states in O^{6+} without, R_{NI}^r , and with, R_{WI}^r , inclusion of the interaction with the $3\ell n\ell'$ Rydberg series. The branching ratios are averaged over all states with the given LS symmetry. The number of $4\ell 4\ell'$ states in each LS symmetry and (in parenthesis) the number of states above threshold in O^{6+} are given as well. The results of Roncin *et al* (1993) were obtained for N^{5+} and takes into account the interaction with the Rydberg series in two different ways as explained in the text. The 'static' results, R_s^r , should be comparable to our results with the interaction with the $3\ell n\ell'$ series included (taking the difference in atom into account), while the dynamic results, R_d^r include the ATR effect. The data in parenthesis correspond to cases where one or more of the individual close-coupling calculations have not converged. These values should be considered as lower limits to the true values. Those symmetries for which the number of states above threshold is different in O^{6+} and in N^{5+} , are indicated by an asterisk.

LS	States	O^{6+} $R_{NI}^r\%$	Present work $R_{WI}^r\%$	N^{5+} $R_s^r\%$	Roncin <i>et al</i> (1993) $R_d^r\%$
$1S^e$	4(1)	3.5	21.2	(8.5)*	(14.3)*
$1P^o$	3(1)	3.0	13.8	2.6	(10.2)
$3P^o$	3	18.6	22.1		
$3P^e$	3(1)	22.3	39.2		
$1D^e$	5(1)	2.5	17.3	17.4	(12.9)
$3D^e$	2	19.5	33.3		
$1D^o$	2	61.9	64.2	39	61.5
$3D^o$	2	5.9	34.9		
$1F^o$	3(1)	0.5	12.6	2.3	8
$3F^o$	3	20.0	36.4		
$1F^e$	1	28.0	53.4	16	75
$3F^e$	3	4.9	22.4		
$1G^e$	3	1.0	27.3	(13.5)*	(10.9)*
$3G^e$	1	12.2	15.3		
$1G^o$	1	9.6	27.9	3.5	28
$3G^o$	1	3.2	31.3		
$1H^o$	1	0.5	38.0	(15)*	2.1*
$3H^o$	1	2.5	5.6		
$3H^e$	1	14.7	72.6		
$1I^o$	1	0.6	54.5	(29)	4

that, insofar as a $4\ell 4\ell'$ term can be identified, the mixing with the $3\ell n\ell'$ Rydberg series does not guarantee a decrease in the autoionization rate. For example, for the highest $3P^o$ term the autoionization rate increases from $2.80 \times 10^{11} \text{ s}^{-1}$, when the interaction with the $3\ell n\ell'$ series is neglected, to $9.63 \times 10^{11} \text{ s}^{-1}$ when it is taken into account.

In the third category, the 31 states lying below the $3\ell 18\ell'$ states can also interact strongly with the lower members of the Rydberg series but they do not show a resonance-type behaviour. The two lowest $1G^e$ terms are good examples (figure 3). Again we find that the inclusion of the interaction with the Rydberg series can lead to an increase in the autoionization rates for individual terms. Examples are the first and the third $3F^o$ terms.

However, considering the average values for each symmetry, the effect of the interaction is in all cases an increase in the stabilization ratio. In table 3, the average radiative branching ratios for the states with a particular LS symmetry are given without, R_{NI}^r , and with, R_{WI}^r , the interaction with the $3\ell n\ell'$ series included. The latter calculation assumes, as mentioned, that the terms are populated according to the amount of $4\ell 4\ell'$ character in each eigenvector. The reason for the increase in the stabilization ratio is the cancellation in the autoionization

rate due to the mixing between $4\ell 4\ell'$ and $3\ell n\ell'$ particularly for those $4\ell 4\ell'$ terms that have a strong interaction with the Rydberg series. For $^1\text{I}^e$ (figure 1) the broad minimum in the autoionization rate increases the stabilization ratio from 0.6 to 54.5%, an increase of nearly a factor of 100!

Roncin *et al* (1993) report two sets of results for the singlet terms in N^{5+} . Both sets are based on close-coupling calculations, the first, R_s^r , is a 'static' calculation in the sense that the distance between the $4\ell 4\ell'$ term and the $N = 3$ threshold is fixed at the free ion value at all times while the second, R_d^r , includes the ATR effect. Both sets of results are shown in table 3. In both calculations a pure $4\ell 4\ell'$ term is assumed to be populated at $t = 0$ and the mixing with the $3\ell n\ell'$ series considered as a function of time. The R_s^r values could be expected to be comparable to our R_{WI}^r values but in fact Roncin *et al* obtain an average stabilization ratio of 14.6% for the singlet terms, considerably lower than our value of 29.7%. However, we note that close agreement cannot be expected. Firstly, the R_s^r results are obtained for N^{5+} and the values presented in table 3 are therefore based on radiative decay rates which, according to the Z^4 scaling law, are smaller than for O^{6+} , while the autoionization rates for N^{5+} and O^{6+} are expected to be closer to each other (see table 4). Secondly, the neglect of the $4\ell 4\ell'$ radiative decay in the R_s^r results contributes also to the difference, since we obtain a stabilization ratio, R_{NI}^r , of 7.8% when only this decay is included. Finally, but perhaps most importantly, the close-coupling calculations neglect the interference between the autoionization decay rates for the $4\ell 4\ell'$ and $3\ell n\ell'$ parts of the wavefunction and this effect is, as mentioned, very important for the degree of stabilization obtained in our calculation.

Considering the rates for particular symmetries, table 3 shows that our R_{WI}^r results in six out of ten cases are closer to the R_d^r than to the R_s^r results. For the symmetries with one or more states above the $N = 3$ threshold, we expected our values to be considerably smaller than those obtained by including the ATR effect since in our approach the states above threshold do not contribute significantly to the stabilization, while the dynamic approach was expected to favour stabilization for these states (Bachau *et al* 1992). In fact, in all cases except two, our R_{WI}^r values are larger than the dynamic R_d^r values. This result is apparently due to the fact that in the close-coupling calculation, the ATR mechanism is so efficient that the $4\ell 4\ell'$ state is transferred into $3\ell n\ell'$ states already at the beginning of the transverse (Roncin 1993). The resulting n values are so low that the final states autoionize instead of stabilize. A good example is provided by the $^1\text{I}^e$ term for which the ATR value is 10 times smaller than ours. In this case the interaction with the series is very strong (figure 1) and the only term for which R_d^r is significantly larger than R_{WI}^r is $^1\text{F}^e$, which has a weak interaction with the Rydberg series. The same effect lies presumably behind the four cases in which the R_s^r values are larger than the R_d^r values.

The average R_{WI}^r ratio is 29.7% for the singlet terms while for the triplet terms the average is as high as 31.2%. The inclusion of the interaction with the Rydberg series increases the stabilization ratios from 7.8% for the singlets and from 12.3% for the triplets. Our overall stabilization ratio for the $4\ell 4\ell'$ manifold is 30.8%.

For the singlet $4\ell 4\ell'$ terms in N^{5+} , Roncin *et al* (1993) obtained an R_s^r value of 14.6%. The inclusion of the dynamic ATR effect in their approach increases the stabilization ratio to 17.7% using a statistical population distribution. Both values are considerably smaller than our static value of 29.7% for O^{6+} , and correspond to an increase of only 3.1% in the stabilization ratio for the singlets in N^{5+} by inclusion of the ATR effect at the appropriate collision velocity.

Table 4. Comparison with other theoretical values for the total autoionization rates (in 10^{12} s^{-1}) of the $4\ell 4\ell'$ terms in O^{6+} . The terms are ordered with the lowest first within each symmetry. Our results correspond to the autoionization rates obtained without, A_{NI}^{a} , and with, A_{WI}^{a} , the interaction with the Rydberg series taken into account. The amount of $4\ell 4\ell'$ character in each term, C_{P} , is shown also. The results due to Vaeck *et al* (1993a) includes the interaction with the Rydberg series but with a limited CI expansion while the results of Ho (1980) are based on the complex rotation method. The differences between Ho's results for the terms below the $N = 3$ threshold and the other calculations are discussed in the text. The results due to Bachau (1984) and Chetoui *et al* (1990) are Feshbach-type calculations. This is also the case for the AIS results from Roncin *et al* (1993) for N^{5+} which correspond to the autoionization rates for the decay to the 2ℓ limits while the results labelled T are the sum of the autoionization rate and the 'auto transfer' rate. For terms above the $N = 3$ threshold, this last result corresponds to the sum of the autoionization rates to the 2ℓ and 3ℓ limits and can therefore be compared with the results of the present work, while this is not the case for the states below threshold (see text).

	O ⁶⁺						N ⁵⁺		
4ℓ4ℓ'	This work A _{NI} ^a	This work C _P (%)	This work A _{WI} ^a	Vaeck <i>et al</i> (1993a)	Ho (1980)	Bachau (1984)	Chetoui <i>et al</i> (1990)	Roncin <i>et al</i> (1993) AIS	Roncin <i>et al</i> (1993) T
¹ S ^e	25.9 53.7 3.59 4.04 ^{b,c}	57 54 9	19.3 43.7 0.108 4.04 ^c		120 641 951 4.96 ^c	31.3 61.6 4.22	25.8 53.9 4.04 1.47–2.84 ^{c,d}	25.2 52.1 4.22 —	133 537 831 ^a 0.628
¹ P ^a	59.3 2.53 127 ^{b,c}	36 25	39.8 1.91		310 878 161 ^c	71.6 2.33	62.8 1.77	62.0 1.45	310 773
¹ D ^e	21.7 38.2 7.55 2.69 162 ^{b,c}	87 59 51 21	17.4 35.9 5.70 0.277 162 ^c			27.4 44.4 8.8 ^c	22.0 40.0 7.46 2.64 171 ^c	23.1 37.4 6.41 2.48 —	
¹ D ^a	0.987 0.0067	84 90	1.02 0.0080				0.951 0.007 52	1.14 0.001 58	
¹ F ^a	26.6 19.7 438 ^{b,c}	61 36	31.8 12.0				25.9 22.1 756 ^c	24.7 18.4 —	
¹ F ^e	0.888	55	0.343				0.0266	0.868	399
¹ G ^e	49.2 9.01 25.5	67 29 3	33.4 1.11 0.297	68.9 4.20 38.6			48.1 6.90 24.1	49.2 8.56 23.4	
¹ G ^a	1.36	76	1.60				1.12	1.40	
¹ H ^a	31.6	8	1.24				29.5	29.1	
¹ I ^e	13.2	7	0.238				6.08	12.2	
³ P ^a	14.3 3.89 0.280	85 83 19	4.76 6.16 0.963		107 316 88.9				

^a This term is above the $N = 3$ threshold in N^{5+} but not in O^{6+} .

^b This term is above the $N = 3$ threshold and interacts weakly with the Rydberg series, thus $A_{\text{NI}}^{\text{a}} = A_{\text{WI}}^{\text{a}}$.

^c Decay to $N = 3$ included.

^d The two values represent a lower and an upper limit, respectively.

^e Due to convergence problems these numbers were expected to be less accurate.

3.2. Comparison with other theoretical work

For the $4\ell 4\ell'$ configurations some earlier work concerning autoionization rates has been published and in table 4 we compare our autoionization rates with previous results. Two

sets of values are shown. One set, A_{NI}^{a} , for which the interaction with the Rydberg series has been neglected and another, A_{WI}^{a} , for which it has been included. We have already pointed out that when the mixing is strong, the choice of a particular state as being '4ℓ4ℓ' SL' can be rather arbitrary. Column 3 shows the total amount of 4ℓ4ℓ' for each term and we note that in six cases the maximum 4ℓ4ℓ' weight is below 25%. This should be kept in mind in the comparisons. Our earlier calculations (Vaeck *et al* 1993a) for the $^1\text{G}^{\circ}$ terms were made with limited CI expansions which were not expected to be adequate for the highest of the three terms. For the other two, the agreement with the new results is satisfactory.

The Feshbach formalism has been used by Bachau (1984) and by Chetoui *et al* (1990) to calculate positions and autoionization widths for the 4ℓ4ℓ' states in O^{6+} . In the former calculation configurations of the type 4ℓ $n\ell'$ ($4 \leq n \leq 7$) were included while the latter calculation with a much larger basis included the first members of the Rydberg series 3ℓ $n\ell'$ (n up to 9) as well. The two calculations give comparable results; the main differences can be attributed to the larger basis set used by Chetoui *et al* (1990). These results can be compared with our values with the interaction with the 3ℓ $n\ell'$ states neglected (A_{NI}^{a}). We notice a better agreement with the results of Chetoui *et al* (1990) which shows that the effect of the inclusion of the first members of the Rydberg series on the autoionization rates is small.

The autoionization rates vary relatively little along the isoelectronic sequence. Roncin *et al* (1993) applied the Feshbach formalism to the calculation of the autoionization rates for the 4ℓ4ℓ' singlet terms in N^{5+} . Their results (labelled AIS), neglecting the interaction with the 3ℓ $n\ell'$ series, are in very good agreement with ours at the same level of approximation (A_{NI}^{a}) despite the different atoms involved. In the dynamic approach of Roncin *et al* (1993), the 3ℓ $n\ell'$ Rydberg states (for $n \rightarrow \infty$) are considered as a 'low continuum' into which the 4ℓ4ℓ' states 'auto transfer' with a characteristic 'width'. The sum (labelled T) of this auto transfer width and the autoionization width to the 2ℓ limits is also given in table 4 for the states above the $N = 3$ threshold as well as for all the $^1\text{S}^{\circ}$ and $^1\text{P}^{\circ}$ terms. For the states above threshold these values can be compared to our A_{WI}^{a} results, while for the states below the $N = 3$ threshold, they cannot, since here the mixing between the 4ℓ4ℓ' states and the Rydberg series is not an autoionization process but a transfer of 4ℓ4ℓ' character to the 3ℓ $n\ell'$ states. Bachau *et al* (1993) noticed, however, that the T values are in rather good agreement with results of Ho (1987) for the singlet 4ℓ4ℓ' manifold in N^{5+} . Ho (1980) calculated energy and autoionization widths for the 4ℓ4ℓ' $^1\text{S}^{\circ}$, $^1\text{P}^{\circ}$ and $^3\text{P}^{\circ}$ terms in the two-electron ions N^{5+} and O^{6+} using the complex coordinate method with Hylleraas basis sets and has later extended his results to all symmetries for N^{5+} (Ho 1987). His results for O^{6+} are given in table 4 and are one or two orders of magnitude larger than our results for the same ion. However, the agreement between the results of Ho (1980, 1987) for N^{5+} and the 'total' width of Roncin *et al* (1993) is very good. As pointed out by Bachau *et al* (1993) this observation raises the question of the interpretation of the results of the complex coordinate method for states overlapping with Rydberg series.

3.3. The 3ℓ $n\ell'$ series and the scaling laws

It is generally assumed that the autoionization decay rate along a Rydberg series decreases as n^{-3} while the radiative decay rates stay relatively constant. This assumption implies that from some n value radiative decay will become dominant. We show in figure 4 the autoionization rates for the six unperturbed 3ℓ $n\ell'$ $^1\text{G}^{\circ}$ Rydberg series (3sng, 3pnf, 3pnh, 3dnd, 3dng and 3dni) as a function of n . Although for large n the n^{-3} behaviour is observed, the first members of the series do generally not follow the same trend. This

Figure 4. Autoionization rates, $A^3 \equiv A_{\text{NI}}^3$ in au, of the six $3\ell n\ell' \ ^1G^\circ$ Rydberg series as function of the principal quantum number n . Only the interaction between the series has been taken into account. The average radiative rate $3.96 \times 10^{11} \text{ s}^{-1}$ for the Rydberg series of this symmetry is indicated by the broken line. The logarithmic horizontal scale is chosen to emphasize the n^{-3} behaviour of the decay rates.

is important because it is common practise to use the n^{-3} dependence to extrapolate the autoionization rates to the high members of a Rydberg series. With the radiative decay estimated as $3.96 \times 10^{11} \text{ s}^{-1}$ (the average radiative decay rate of the $3\ell 14\ell' \ ^1G^\circ$ states according to table 2) it is only for n approaching 40 that radiative decay becomes the dominant de-excitation mechanism for *all* series. Roncin *et al* (1993) have characterized the competition between autoionization and fluorescence in a given $3\ell n\ell'$ series by the value $\tilde{n}$ which corresponds to the n value for which autoionization and radiative rates are equal. For N^{5+} , using hydrogenic radiative decay rates and by scaling the autoionization rates from the $3\ell 5\ell'$ states (Bachau *et al* 1991), Roncin *et al* (1993) found that two-thirds of the Rydberg series have $\tilde{n} \leq 20$ while the mean value of $\tilde{n}$ lies between 20 and 25. This is in reasonable agreement with the results presented in figure 4 for the autoionization widths of the unperturbed $^1G^\circ$ series. For $n = 23$, four of the six series have an autoionization rate smaller than the radiative rate while for $n = 25$ only one series has an autoionization rate larger than the radiative rate. In terms of the close-coupling calculations, reported by Roncin *et al* (1993), this means that the $4\ell 4\ell'$ amplitude, which is transferred into $3\ell n\ell'$ states with $n \leq \tilde{n}$, decay by autoionization and thus does not contribute to stabilization.

We show in figure 5 autoionization decay rates of the same six $^1G^\circ$ series but including this time the interaction with the $4\ell 4\ell'$ states. It is clear from this figure that the mixing of the $4\ell 4\ell'$ states with the many interacting series results in a very irregular behaviour of the autoionization rates while not changing the average values very much. Two thirds of the series stabilize, as before, at $n = 22$, and similarly all series stabilize around $n = 40$ and the main difference is that the n value above which one series is still preferentially autoionizing is somewhat larger than before. Thus the effect of the interaction with the $4\ell 4\ell'$ states is on average a slight *increase* in the autoionization rate of the $3\ell n\ell'$ series, while, as mentioned before, the effect on the $4\ell 4\ell'$ states, on the average, is a clear decrease in the autoionization rate.

Figure 5. Autoionization rates, $A^3 \equiv A_{WI}^3$ in au, of the six $3\ell n\ell'$ $^1G^\circ$ Rydberg series as a function of the principal quantum number n . The interaction with the three $4\ell 4\ell'$ $^1G^\circ$ terms as well as the interaction between the series has been taken into account. The average radiative rate $3.96 \times 10^{11} \text{ s}^{-1}$ for the Rydberg series of this symmetry is indicated by the broken line. The logarithmic horizontal scale is chosen to emphasize the n^{-3} behaviour of the decay rates.

3.4. The $4\ell 5\ell'$ configurations

The $4\ell 5\ell'$ terms in O^{6+} are located more than 11 eV above the $N = 3$ threshold and we have estimated that in a molecular approach to the collision dynamics they do not cross the threshold (Vaeck and Hansen 1993). Therefore, we assume that the decay properties of the $4\ell 5\ell'$ states can be described unambiguously using a static approach.

In table 5, results for energies, autoionization and radiative decay rates and fluorescence yields for the 120 $4\ell 5\ell'$ terms in O^{6+} are shown. Similarly to the $4\ell 4\ell'$ states, the $4\ell 5\ell'$ states are strongly mixed within the Layzer complex and we give in column 2 the main components of the eigenvector for each term in the hydrogenic (B -spline) basis.

Chen and Lin (1993) have calculated fluorescence yields using a CI approach with a hydrogenic basis set for singlet terms in the $4\ell 4\ell'$, $5\ell 5\ell'$ and $6\ell 6\ell'$ as well as the $5\ell 6\ell'$ manifold in Ar^{16+} . Due to the Z^4 scaling of the radiative decay rates the radiative branching ratios are expected to be larger in Ar^{16+} than in O^{6+} . For the $4\ell 4\ell'$ states, which in Ar^{16+} lie considerably below the $N = 3$ threshold, Chen and Lin (1993) find that more than half of the singlets have a radiative branching ratio larger than 40% and two states have a fluorescence yield of nearly 100%. Indeed, the closest available limit (the $N = 2$ limit) is more than 550 eV below the first $4\ell 4\ell'$ state. The calculations included intrashell correlation only, i.e. interaction between the states in the complexes. Adjacent Rydberg series were not considered. For the higher configurations for which additional limits become available, Chen and Lin observed a large reduction of the fluorescence yield in agreement with the previous results of Gou *et al* (1991). For the $5\ell 5\ell'$ manifold, only five states were calculated to have a radiative branching ratio larger than 20% and for the $6\ell 6\ell'$ manifold most of the states have fluorescence yields smaller than 10% with the closer limits being the $N = 3$ limit at approximately 160 eV and the $N = 4$ limit at about 80 eV, respectively. Chen and Lin classified the terms according to the $(K, T)^A$ scheme where K , T and A are quantum numbers that describe the correlation of the doubly-excited states (Lin 1986). For $n\ell n\ell'$

Table 5. Total energies E , autoionization rates A^* , radiative decay rates A^r and radiative branching ratios R^r for the $4l5l'$ states in O^{6+} . The radiative decay rate is obtained from a CI calculation using the Cowan program (Cowan 1981) as discussed in the text. The states are ordered according to energy within each LS symmetry. For each term the main eigenvector components in the hydrogenic B -spline basis are given.

LS	Main eigenvector components	E (au)	$A^* \times 10^{12}$ (s^{-1})	$A^r \times 10^{12}$ (s^{-1})	R^r (%)
$1S^e$	47% 4s5s 44% 4p5p	-3.111 19	5.0	0.224	0.3
	27% 4s5s 53% 4d5d 11% 4f5f	-3.052 19	393	0.136	0.03
	56% 4f5f 18% 4p5p 13% 4s5s	-2.970 70	535	0.164	0.03
	29% 4d5d 24% 4p5p 19% 4f5f	-2.872 67	2.1	0.230	9.9
$3S^e$	41% 4s5s 47% 4p5p 11% 4d5d	-3.131 30	2.7	0.256	8.7
	29% 4s5s 53% 4d5d 15% 4f5f	-3.093 15	10.4	0.111	1.1
	18% 4s5s 21% 4p5p 59% 4f5f	-3.035 29	17.5	0.153	0.9
	11% 4s5s 28% 4p5p 33% 4d5d 22% 4f5f	-2.940 15	0.15	0.226	60
$1P^o$	33% 4s5p 33% 5s4p 15% 4p5d 15% 5p4d	-3.129 00	3.8	0.264	6.5
	17% 4s5p 20% 5p4d 20% 4d5f 22% 5d4f	-3.088 83	12.7	0.207	1.6
	14% 4s5p 22% 5s4p 29% 4p5d 22% 5p4d	-3.083 08	203	0.271	0.1
	19% 4p5d 16% 5p4d 30% 5d4f 15% 4f5g	-3.025 81	10.4	0.185	1.7
	13% 4s5p 17% 5s4p 27% 4d5f 25% 5d4f	-3.009 14	587	0.169	0.03
	58% 4f5g	-2.909 45	14.1	0.096	0.7
	12% 4s5p 22% 4p5d 12% 5p4d 20% 4d5f	-2.904 18	50.1	0.247	0.5
$3P^o$	12% 4s5p 18% 5s4p 26% 4p5d 30% 5p4d	-3.113 12	6.77	0.264	3.8
	38% 4s5p 33% 5s4p 15% 4p5d 10% 5p4d	-3.108 47	74.2	0.252	0.3
	16% 4s5p 18% 5s4p 27% 4d5f 30% 5d4f	-3.064 81	17.2	0.157	0.9
	17% 4p5d 20% 5p4d 19% 4d5f 17% 5d4f	-3.054 25	198	0.229	0.1
	13% 4s5p 14% 5s4p 48% 4f5g	-2.988 11	9.43	0.159	1.7
	23% 5p4d 16% 4d5f 36% 5d4f	-2.978 98	40.0	0.185	0.5
	15% 4p5d 25% 4d5f 37% 4f5g	-2.926 96	0.38	0.173	31
$1P^e$	57% 4p5p 39% 4d5d	-3.11339	5.70	0.335	5.6
	33% 4p5p 25% 4d5d 41% 4f5f	-3.066 54	10.7	0.228	2.1
	35% 4d5d 53% 4f5f	-2.996 96	0.18	0.148	45
$3P^e$	63% 4p5p 32% 4d5d	-3.085 96	141	0.360	0.3
	27% 4p5p 33% 4d5d 34% 4f5f	-3.024 01	207	0.210	0.1
	30% 4d5d 55% 4f5f	-2.955 43	0.95	0.131	12
$1D^e$	26% 4s5d 36% 5s4d 18% 4p5f 17% 5p4f	-3.106 32	8.06	0.157	1.9
	24% 4s5d 16% 5s4d 45% 4p5p	-3.101 22	98.8	0.281	0.3
	12% 4s5d 61% 4d5d 12% 4f5f	-3.056 64	236	0.141	0.06
	14% 4s5d 18% 5s4d 32% 5p4f 20% 4d5g	-3.049 14	9.55	0.186	1.9
	24% 4p5p 27% 4p5f 28% 5p4f	-3.037 72	301	0.291	0.1
	70% 4f5f	-2.989 90	222	0.111	0.05
	11% 5s4d 10% 4p5p 10% 5p4f 41% 4d5g	-2.940 88	97.1	0.173	0.2
	13% 4s5d 22% 4p5f 11% 4d5d 17% 4d5g	-2.914 14	30.6	0.231	0.7
$3D^e$	21% 4s5d 20% 5s4d 42% 4p5p	-3.124 06	4.30	0.266	5.8
	52% 4d5d 13% 4f5f	-3.092 73	9.43	0.172	1.8
	12% 4s5d 11% 5s4d 15% 4p5p 39% 5p4f	-3.078 79	47.6	0.276	0.6
	19% 4s5d 25% 5s4d 34% 4p5f	-3.076 45	105	0.138	0.1
	59% 4f5f	-3.036 07	11.7	0.120	1.0
	13% 4s5d 14% 5s4d 38% 4p5f	-3.005 69	44.4	0.155	0.3
	14% 4p5p 15% 4d5d 35% 4d5g 16% 4f5f	-2.997 85	1.48	0.214	13
	11% 4s5d 25% 4p5f 35% 4d5g	-2.948 68	5.29	0.210	3.8

Table 5. continued.

<i>LS</i>	Main eigenvector components	<i>E</i> (au)	<i>A</i> ^a × 10 ¹² (s ⁻¹)	<i>A</i> ^r × 10 ¹² (s ⁻¹)	<i>R</i> ^r (%)
¹ D ^o	29% 4p5d 31% 5p4d 21% 4d5f 17% 5d4f	-3.092 67	13.3	0.236	1.7
	40% 4p5d 42% 5p4d	-3.080 51	90.1	0.276	0.3
	19% 4p5d 20% 4d5f 15% 5p4d 20% 5d4f 24% 4f5g-3.035 58	6.45	0.172	2.6	
	27% 4d5f 49% 5d4f	-3.022 10	15.9	0.138	0.9
	21% 4d5f 68% 4f5g	-2.978 35	0.010	0.098	91
³ D ^o	39% 4p5d 39% 5p4d 11% 4d5f	-3.107 22	59.9	0.273	0.5
	39% 4p5d 24% 5p4d 27% 5d4f	-3.057 46	185	0.156	0.08
	19% 5p4d 40% 4d5f 26% 5d4f	-3.054 46	15.2	0.245	1.6
	14% 4p5d 15% 5p4d 22% 4d5f 33% 5d4f	-2.989 74	80.6	0.158	0.2
	17% 4d5f 76% 4f5g	-2.957 71	1.17	0.103	8.1
¹ F ^o	10% 4s5f 39% 4p5d 37% 5p4d	-3.115 68	6.73	0.254	3.6
	20% 4s5f 18% 5s4f 24% 4d5f 28% 5d4f	-3.079 43	5.15	0.100	1.9
	15% 4s5f 50% 5s4f 20% 4p5d	-3.060 77	142	0.174	0.1
	20% 4s5f 16% 5p4d 20% 4p5g 19% 4d5f 14% 5d4f-3.059 08	42.9	0.136	0.3	
	33% 4d5f 37% 5d4f	-3.027 83	321	0.144	0.04
	36% 4p5g 44% 4f5g	-2.998 29	68.7	0.167	0.2
	20% 5p4d 13% 4p5d 28% 4f5g	-2.951 52	223	0.190	0.09
	16% 4s5f 13% 4p5d 21% 4p5g 15% 4d5f 15% 4f5g-2.920 82	29.1	0.211	0.7	
³ F ^o	26% 4s5f 32% 5s4f 22% 5p4d	-3.094 35	9.53	0.142	1.5
	11% 4s5f 43% 4p5d 33% 5p4d	-3.090 52	82.3	0.260	0.3
	35% 4d5f 29% 5d4f	-3.070 53	12.1	0.126	1.0
	14% 4s5f 18% 5s4f 26% 4d5f 35% 5d4f	-3.051 02	40.0	0.098	0.2
	12% 4s5f 19% 5s4f 19% 4p5d 18% 4p5g 12% 4d5f-3.025 61	8.92	0.223	2.4	
	14% 5s4f 25% 5p4d 13% 5d4f 32% 4f5g	-3.018 71	7.16	0.127	1.7
	36% 4p5g 39% 4f5g	-3.008 26	13.8	0.169	1.2
	17% 4s5f 15% 4p5d 26% 4p5g 13% 4d5f 13% 4f5g-2.967 81	10.5	0.207	1.9	
¹ F ^e	19% 4p5f 18% 5p4f 54% 4d5d	-3.096 52	5.49	0.202	3.5
	25% 4p5f 20% 5p4f 20% 4d5d 27% 4f5f	-3.070 59	10.4	0.189	1.8
	37% 4p5f 56% 5p4f	-3.050 21	26.4	0.241	0.9
	20% 4d5d 15% 4d5g 60% 4f5f	-3.030 74	0.031	0.108	78
	16% 4p5f 69% 4d5g	-3.006 48	8.73	0.159	1.8
³ F ^e	43% 4p5f 42% 5p4f 11% 4d5g	-3.079 09	3.91	0.264	6.3
	15% 4p5f 27% 5p4f 51% 4d5d	-3.069 73	61.5	0.183	0.3
	24% 4p5f 19% 5p4f 19% 4d5d 33% 4f5f	-3.031 26	176.	0.182	0.1
	13% 4d5d 52% 4d5g 20% 4f5f	-3.011 35	18.0	0.128	0.7
	12% 4p5f 12% 4d5d 32% 4d5g 38% 4f5f	-2.979 93	47.9	0.142	0.3
¹ G ^e	41% 4p5f 41% 4d5d	-3.073 46	19.9	0.245	1.2
	16% 4p5f 61% 5p4f	-3.072 74	82.6	0.182	0.2
	40% 4s5g 44% 4d5g	-3.042 48	119	0.080	0.07
	12% 4s5g 72% 4f5f	-3.013 54	264	0.100	0.04
	24% 5p4f 26% 4d5d 26% 4d5g	-2.981 61	388	0.140	0.04
	16% 4s5g 26% 4p5f 15% 4d5d 17% 4d5g 10% 4f5f-2.932 12	75.2	0.202	0.3	
³ G ^e	26% 4p5f 19% 5p4f 49% 4d5d	-3.102 12	9.68	0.218	2.2
	15% 4p5f 64% 4f5f	-3.058 64	11.4	0.115	1.0
	28% 4s5g 35% 5p4f 21% 4d5g	-3.045 98	13.6	0.093	0.7
	21% 4p5f 22% 5p4f 45% 4d5g	-3.038 56	21.5	0.201	0.9
	26% 4s5g 15% 5p4f 24% 4d5d 23% 4f5f	-3.023 72	0.88	0.139	14
	23% 4s5g 28% 4p5f 14% 4d5d 22% 4d5g	-2.989 45	24.1	0.178	0.7

Table 5. continued

<i>LS</i>	Main eigenvector components	<i>E</i> (au)	$A^a \times 10^{12}$ (s ⁻¹)	$A^r \times 10^{12}$ (s ⁻¹)	<i>R^r</i> (%)
¹ G ^o	13% 4p5g 74% 4d5f	-3.059 82	4.23	0.148	3.4
	87% 5d4f	-3.053 80	8.35	0.173	2.0
	71% 4p5g 15% 4f5g	-3.040 95	17.4	0.196	1.1
	18% 4d5f 66% 4f5g	-3.014 77	11.5	0.105	0.9
³ G ^o	47% 4d5f 45% 5d4f	-3.078 70	3.31	0.133	3.9
	77% 4p5g 14% 5d4f	-3.044 03	38.7	0.280	0.7
	36% 4d5f 37% 5d4f 14% 4f5g	-3.009 82	173	0.114	0.07
	12% 4d5f 74% 4f5g	-2.975 22	22.6	0.112	0.5
¹ H ^o	49% 4d5f 39% 5d4f	-3.079 47	23.0	0.143	0.6
	50% 4p5g 20% 5d4f 23% 4f5g	-3.024 00	207	0.197	0.1
	15% 4d5f 28% 5d4f 47% 4f5g	-2.978 50	542	0.096	0.02
	30% 4p5g 28% 4d5f 22% 4f5g	-2.938 55	86.9	0.204	0.2
³ H ^o	10% 4p5g 55% 4d5f 26% 5d4f	-3.055 61	22.5	0.137	0.6
	36% 5d4f 56% 4f5g	-3.044 07	26.0	0.093	0.4
	48% 4p5g 25% 5d4f 22% 4f5g	-3.032 29	13.6	0.200	1.4
	37% 4p5g 33% 4d5f 18% 4f5g	-3.007 80	42.2	0.181	0.4
¹ H ^e	93% 4f5f	-3.049 00	7.65	0.084	1.1
	93% 4d5g	-3.036 01	0.446	0.109	20
³ H ^e	51% 4d5g 45% 4f5f	-3.039 16	136	0.094	0.07
	45% 4d5g 50% 4f5f	-2.993 16	100	0.102	0.1
¹ I ^o	37% 4d5g 54% 4f5f	-3.018 16	883	0.077	0.01
	52% 4d5g 36% 4f5f	-2.949 61	240	0.112	0.05
³ I ^o	19% 4d5g 78% 4f5f	-3.040 47	8.68	0.093	1.1
	78% 4d5g 19% 4f5f	-3.026 41	69.0	0.096	0.1
¹ I ^e	98% 4f5g	-3.041 69	43.9	0.055	0.1
³ I ^e	94% 4f5g	-2.982 27	81.9	0.074	0.09
¹ K ^o	87% 4f5g	-2.948 55	295	0.071	0.02
³ K ^o	98% 4f5g	-2.987 08	123	0.049	0.04

states, *A* is always equal to +1, while for *nℓn'ℓ'* states the values *A* = 0 and *A* = -1 are possible also. Chen and Lin found for the singlet terms in the 5ℓ6ℓ' manifold that the autoionization rates are much smaller for the states with *A* = -1 or 0 than for states with *A* = +1. Therefore for *nℓn'ℓ'* configurations a large number of states was expected by Chen and Lin to have a large fluorescence yield. For the singlet 5ℓ6ℓ' terms in Ar¹⁶⁺ slightly less than half the states have a radiative branching ratio larger than 40%. We note, however, that the 5ℓ6ℓ' states in Ar¹⁶⁺ are lying just below the *N* = 4 threshold about 200 eV from the *N* = 3 limit. In contrast, the 4ℓ5ℓ' states in O⁶⁺ are lying above but fairly close to the *N* = 3 threshold and our results show a very small radiative branching ratio for these states. Only four of the 120 states have a radiative branching ratio of more than 40% while 69 states have a radiative branching ratio of less than one per cent. This observation in combination with the very high percentage of radiative stabilization obtained for the 4ℓ4ℓ' manifold seems inconsistent with the (*K*, *T*)^A classification. The reason for the discrepancy is probably that the 4ℓ5ℓ' terms can decay to the *N* = 3 limit while the

majority of the $4\ell 4\ell'$ terms cannot. Even in the absence of overlap with the $3\ell n\ell'$ series, the average stabilization ratio for the $4\ell 4\ell'$ manifold (7.8% for the singlets) is larger than the ratio obtained for the $4\ell 5\ell'$ manifold. Finally we note that Chen and Lin (1993) assume that the neglect of the interaction with adjacent Rydberg series has little influence on the average fluorescence yields. We have not confirmed this assumption and it would be interesting to check its validity for the $5\ell n\ell'$ terms in Ar^{16+} which overlap the $4\ell n\ell'$ series.

Table 6. Autoionization rates for those $4\ell 4\ell'$ terms in O^{6+} that lie above the $N = 3$ limit. The third column shows the partial rates to the $1s$ and 2ℓ limits, $A_{1,2}^a$, the fourth column gives the total rate, $A_{1,2,3}^a$, i.e. including also the contribution from the decay to the 3ℓ limits. The composition of the states are indicated in the second column.

LS	$4\ell 4\ell'$ composition	$A_{1,2}^a 10^{12} \text{ (s}^{-1}\text{)}$	$A_{1,2,3}^a 10^{12} \text{ (s}^{-1}\text{)}$
$^1S^e$	22% $4p^2$ 32% $4d^2$ 33% $4f^2$	0.624	4.04
$^1P^o$	24% $4s4p$ 38% $4p4d$ 31% $4d4f$	6.72	127
$^3P^e$	26% $4d^2$ 67% $4f^2$	0.038	1.75
$^1D^e$	23% $4s^2$ 17% $4p^2$ 20% $4p4f$ 18% $4d^2$ 16% $4f^2$	6.46	162
$^1F^o$	19% $4s4f$ 48% $4p4d$ 28% $4d4f$	13.2	438

The importance of the distance to the nearest limit can be seen from table 6, which shows the decay rates for the five $4\ell 4\ell'$ terms that lie above the $N = 3$ limit in O^{6+} . The autoionization rates to the $N = 1$ and 2 limits and the total rates (including the contribution from the decay to $N = 3$) are shown and it can be seen that the contribution from the decay to the $N = 3$ limits increases the autoionization rates by factors varying between 6.5 and 46. This increase obviously has a very large effect on the degree of stabilization. As mentioned earlier, for the terms located close to the $N = 3$ limit interchannel coupling might be important for the total autoionization rates. However, the effect on the stabilization ratios is small since autoionization predominates. Thus the distance to the closest limit appears to remain a good indication of the strength of the autoionization (Hansen 1989) but a calculation for the $4\ell 5\ell'$ manifold in Ar^{16+} would clearly be worthwhile.

Finally, in table 7 we give the average stabilization ratios for each LS symmetry in the $4\ell 5\ell'$ manifold. The dispersion of the results is much smaller than for the $4\ell 4\ell'$ states. All symmetries have an average stabilization ratio smaller than 10% except $^3S^e$, $^1P^e$, $^1D^o$, $^1P^e$ and $^1H^e$ which have stabilization ratios between 11 and 19%. Among these, the singlets have unnatural parity. In contrast to the situation for the $4\ell 4\ell'$ terms, the singlets in $4\ell 5\ell'$ have a larger average stabilization ratio (4.2%) than the triplets (2.0%). The overall stabilization ratio for the $4\ell 5\ell'$ manifold, 2.6%, is more than 11 times smaller than for the $4\ell 4\ell'$ states.

4. Discussion

The overall stabilization ratio for the $4\ell 4\ell'$ manifold, obtained including the interaction with the $3\ell n\ell'$ Rydberg series, is equal to 30.8% assuming a statistical population. For the $4\ell 5\ell'$ manifold the calculated ratio is 2.6%. The 25% TDC found experimentally by Roncin *et al* (1991) and Gaboriaud *et al* (1993) for the collision system $\text{O}^{8+} + \text{Ar}$, has been broken down into an overall stabilization ratio of $59 \pm 9\%$ for the $4\ell 4\ell'$ manifold and of $11 \pm 1\%$ for the $4\ell 5\ell'$ manifold (Gaboriaud *et al* 1993). Assuming a statistical population of the states it is clear that the results we have obtained cannot explain the experimental results.

Table 7. Branching ratios for radiative decay for the $4\ell 5\ell'$ states. The R_{av}^r ratios are averaged over all states with the same LS symmetry. The number of states in each LS symmetry is indicated also.

LS	States	R_{av}^r %
$^1S^e$	4	2.6
$^3S^e$	4	18
$^1P^o$	7	1.6
$^3P^o$	7	5.5
$^1P^e$	3	18
$^3P^e$	3	4.1
$^1D^e$	8	0.7
$^3D^e$	8	3.3
$^1D^o$	5	19
$^3D^o$	5	2.1
$^1F^o$	8	0.9
$^3F^o$	8	1.3
$^1F^e$	5	17
$^3F^e$	5	1.5
$^1G^e$	6	0.3
$^3G^e$	6	3.3
$^1G^o$	4	1.9
$^3G^o$	4	1.3
$^1H^o$	4	0.2
$^3H^o$	4	0.7
$^1H^e$	2	11
$^3H^e$	2	0.1
$^1I^o$	2	0.0
$^3I^o$	2	0.6
$^1I^e$	1	0.1
$^3I^e$	1	0.1
$^1K^o$	1	0.0
$^3K^o$	1	0.0

Similarly, under the same assumption the calculations including the ATR effect by Roncin *et al* (1993) for the $4\ell 4\ell'$ singlet terms in N^{5+} give a stabilization ratio of 17.7%. However, also in N^{5+} a much larger stabilization ratio ($>52\%$) is necessary to explain the observed TDC probabilities. A prime suspect for the disagreement is of course the assumption of a statistical population distribution.

In this section we briefly recall the assumptions concerning the states which are populated in the double capture. Two types of assumptions are involved. Firstly, assuming that $4\ell 4\ell'$ and not $3\ell n\ell'$ states are populated in $O^{8+} + Ar$ collisions and secondly assuming that the population distribution over the $4\ell 4\ell'$ manifold is statistical in origin. Both assumptions can be questioned. The best confirmed property of the ECBM is that the possible final states lie in an energy range characterized as the 'reaction window' (Mack 1987) and it appears fairly well established that if a $n\ell n'\ell'$ configuration with $n \approx n'$ appears in the middle of the reaction window, this configuration will be strongly favoured (Barat *et al* 1993). In the present case, this is the $4\ell 5\ell'$ configuration in agreement with the observed population of 71% for this configuration (Gaboriaud *et al* 1993). In fact the $4\ell 4\ell'$ configuration is lying outside the reaction window and the population mechanism might be more complicated. The necessity of using Ar as a collision partner in order to populate the $4\ell 4\ell'$ manifold adds to the uncertainty about the pick-up mechanism. Martin *et al* (1992) and Ali *et al* (1993) have

found evidence for the population of Rydberg states in $\text{Kr}^{q+} + \text{Kr}$ and $\text{Ar}^{q+} + \text{Ar}$ collisions for large values of q and Vaecck *et al* (1993b) have drawn attention to the observation of $2\ell n\ell'$ Rydberg states in collisions between, for example, C^{6+} and He (Stolterfoht *et al* 1990) and noted that a similar process, which does not involve overlap with a Rydberg series in the free atom, in the present case could lead to considerable amounts of stabilization. Extensive discussions have taken place concerning the origin of the $2\ell n\ell'$ population in, for example, C^{4+} , but in the present context, the important feature is its existence as such. Since the $4\ell 4\ell'$ configuration is degenerate with the $3\ell n\ell'$ Rydberg series it is not possible to distinguish between the two with the help of energy gain measurements and it is clearly difficult to unravel the initial population distribution experimentally. Nevertheless studies concerning this point would be valuable. It is worth mentioning that also the influence on the population mechanism of the mixing between $4\ell 4\ell'$ and $3\ell n\ell'$ states at the point of pick-up requires attention.

The second point concerns the assumption of a statistical population distribution within the $4\ell 4\ell'$ manifold. The dispersion between the results in table 3 shows that the overall stabilization ratio is critically dependent on this assumption and that it is possible by a judicious choice of population distribution to obtain nearly any desired value. While very little is known about the actual population distribution over the $4\ell 4\ell'$ manifold, Bordenave-Montesquieu *et al* (1987) and Posthumus *et al* (1992) have studied the population distribution for the $3\ell 3\ell'$ manifolds in C^{4+} , N^{5+} and O^{6+} . The experimental distributions were found to be non-statistical and, in fact, also velocity dependent. In both studies it was found that particular terms with large L values are preferentially populated and Posthumus *et al* (1992) observed that the population of these states increases with increasing projectile velocity compared to the population of states with small L values. For 24–120 keV C^{6+} colliding with H_2 , the highest angular momentum state ($^1\text{G}^e$) is predominantly populated. At 60 keV it contributes more than 50% of the $3\ell 3\ell'$ population.

On the other hand, in slow collisions between highly charged ions and neutral targets, the total spin of the captured electrons does not change. This means that in collisions with He only singlet terms of O^{6+} can be populated, while in collisions with Ar it is possible to populate both singlet and triplet terms. There are no systematic studies of the relative population of the triplet states in comparison with the population of the singlets but for beryllium-like systems ($\text{N}^{5+} + \text{Ar}$) Mack (1987) found that the triplets have a large population.

The results in table 3 show that the terms with the highest L value for both singlet and triplets have stabilization ratios larger than 50%. If these terms are predominantly populated during the collision, the experimental results can easily be understood. It is interesting to note that if this assumption is correct we can predict an increase in the stabilization ratio with velocity. However, we find that the ECBM augmented to take angular momentum effects into account following the recipe given by Burgdörfer *et al* (1987) at this low energy predicts that predominantly $\ell = 0$ electrons are picked up and the calculated stabilization ratios show that this will not lead to the necessary degree of stabilization. This is perhaps another indication that the pick-up in this case does not follow the conventional ECBM recipe. On the other hand, in the dynamic ATR approach, an increase in the velocity of the projectile decreases the time the $4\ell 4\ell'$ state spends 'within' the Rydberg series and therefore decreases the efficiency of the ATR mechanism. However, the discussion concerning the difference between the R_s^r and R_d^r ratios in table 3 shows that it is not obvious that this always will lead to a reduction of the stabilization ratio and also this mechanism can have a maximum at a velocity larger than zero. For $\text{O}^{8+} + \text{He}$, Wu *et al* (1993b) have observed a velocity dependence with an initial decrease followed by a second maximum. Ali *et al* (1993) have observed a monotonic decrease in the stabilization ratio with velocity for collisions between

Ar^{8+} and Ar and Wu *et al* (1993a) have observed a similar trend in the stabilization ratio for $\text{Ar}^{16+} + \text{He}$ collisions. In the latter case, Wu *et al* noted that the decrease could be explained by the fact that the observed Q values shows that pick-up in the $1s^27\ell7\ell'$ configurations is becoming more probable relative to $1s^26\ell7\ell'$ with increasing velocity, since the $1s^27\ell7\ell'$ configurations, lying just above the $1s^25\ell$ thresholds, are expected to have a smaller stabilization ratio than the $1s^26\ell7\ell'$ configurations (Hansen 1989).

It has been pointed out that there is little evidence for the population of terms with unnatural parity in double capture (Gaboriaud *et al* 1993). This can simply be because very few singlet terms with unnatural parity exist for $n\ell n'\ell'$ configurations with $n \leq 3$ but it could possibly also be because these terms have a high stabilization ratio and therefore are difficult to observe in electron spectra. The $^3\text{H}^e$ term is an example of this type. The average stabilization ratio for the unnatural parity states in O^{6+} is 39% compared to 25% for the natural parity states. In fact, Vaeck and Hansen (1993) have mentioned that a selective population of some of these terms can explain the experimental results.

Theoretical calculations on different levels of sophistication have been carried out to determine the population distribution over the $3\ell3\ell'$ states populated during the collision between O^{8+} and He (Harel and Salin 1981, Posthumus *et al* 1992, Chen and Lin 1991, Chen *et al* 1991 and Harel and Jouin 1992). Studies of this type for the $4\ell4\ell'$ and $3\ell n\ell'$ states are needed in order to understand the experimental results.

5. Conclusions

We have studied the decay properties of the 120 $4\ell5\ell'$ and the 44 $4\ell4\ell'$ terms in O^{6+} using a truncated diagonalization method with B -spline basis sets. The $4\ell4\ell'$ states are lying in the same range of energy as the $3\ell n\ell'$ Rydberg series and we find that the interaction with the Rydberg series increases the total stabilization ratio of the $4\ell4\ell'$ manifold considerably. We compare our results for the $4\ell4\ell'$ and $4\ell5\ell'$ states with the prediction of Chen and Lin (1993) who, using the $(K, T)^A$ classification, proposed that in $n\ell n'\ell'$ configurations, the terms with $A < 1$ can have large stabilization ratios and noted that such states do not exist in $n\ell n\ell'$ configurations. We do not observe this behaviour. We obtain, in fact, a larger radiative branching ratio for the $4\ell4\ell'$ than for the $4\ell5\ell'$ states in contradiction to the $(K, T)^A$ model that, however, neither considers the overlap between $4\ell4\ell'$ and a Rydberg series nor the importance of the proximity of the closest threshold.

The main purpose of this work was to understand the high TDC probability observed by Roncin *et al* (1991) and Gaboriaud *et al* (1993) for the doubly-excited states populated during the collision between O^{8+} and Ar. The 25% TDC obtained experimentally has been interpreted as corresponding to stabilization ratios of 59% and 11% for the $4\ell4\ell'$ and $4\ell5\ell'$ manifolds, respectively (Gaboriaud *et al* 1993). Assuming a statistical population distribution, we find 30.8% and 2.6% stabilization for the $4\ell4\ell'$ and $4\ell5\ell'$ states, respectively. We agree with Roncin *et al* (1993) that the mixing between the $4\ell4\ell'$ states and the high members of the Rydberg series can increase the radiative stabilization although the mechanisms are different. In our case the stabilization ratio is increased from 8% to 30% for the singlets and from 12% to 31% for the triplet terms.

While the average stabilization ratios are too small to explain the experimental data, we find very large stabilization ratios for particular states and particular LS symmetries. Therefore we conclude that in order to compare with the experimental stabilization ratios, information about the population distribution for the $4\ell4\ell'$ and $4\ell5\ell'$ states is required and we suggest a more careful study the formation of these states.

The major computational problem in these calculations is how to reliably describe the

interaction between a perturber and very high-lying Rydberg states and we have adapted our *B-spline methods* (van der Hart and Hansen 1992b, 1993a) to this purpose. The ease with which this has been possible shows the flexibility of the *B-spline approach*. The main remaining uncertainty is connected with the determination of the total radiative decay rates which are difficult to calculate when the initial states are so mixed as is the case here.

Acknowledgments

We thank Drs R Ali, H Bachau, A Bárány, C D Lin, R. Morgenstern, P Roncin, N Stolterfoht and W Wu for providing information about their work prior to publication and/or for stimulating discussions. This work was sponsored by the Stichting Nationale Computerfaciliteiten (National Computing Facilities Foundation, NCF) for the use of supercomputer facilities with financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for Scientific Research, NWO).

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