Variance of the distributions of energy levels and of the transition arrays in atomic spectra. II. Configurations with more than two open subshells

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In a previous article it has been shown how the variance of the distribution of the lines belonging to a transition array between two atomic configurations can be computed exactly. The agreement with experimental unresolved spectral patterns was good. We generalize the formal expansions obtained previously for simple arrays to cases with an arbitrary number of open subshells. The results are expressed in terms of the formulas already published [Phys. Rev. A 20, 2424 (1979)].

I. INTRODUCTION

In the problem of the identification and analysis of the spectra of highly ionized atoms, the *ab initio* evaluation of the variances σ^2 of the transition arrays proves to be a valuable tool.[1-4] Indeed, it often happens that the numerous lines of an array between two configurations are merged together by various line-broadening mechanisms, so that they cannot be seen individually; then, the spectrum shows characteristic "bands," each of which originates in a different ionization state and pair of configurations. The full width at half maximum of such a band, if assumed Gaussian in shape, is equal to $2\sigma\sqrt{2}\ln 2 = 2.355\sigma$. But the mathematical quantity σ^2 does not depend, of course, on any shape assumption.

The formal expansions of σ^2 for the simplest types of arrays, nl^{N+1}-$nl^N n'l'$ and $nl^N n'l'$-$nl^N n''l''$, have been determined by Moszkowski[5] and Bauche-Arnoult *et al.*,[1] and the latter authors have dealt with the case of intermediate coupling and published explicit tables. However, in the course of experimental interpretations of some spectra, there appeared transition arrays whose variances could not be calculated with the formulas already published[1] because they involved configurations with three open subshells, e.g., $3d^9 4s$-$3d^8 4f\,4s$ and $3p^5 3d^{10}$-$3p^5 3d^9 4p$ in Mo XV and Mo XVI.[6] In the present paper, we derive the formulas for the variance of any array where three or more open subshells occur, and for the mean energy (wave number) of such an array, which is also useful in the experimental interpretations.

The variance for the transition array A-B is defined as

$$\sigma^2 = \mu_2 - (\mu_1)^2 ,$$

where

$$\mu_n = \sum_{a,b} [(a \mid H \mid a) - (b \mid H \mid b)]^n w_{ab}/W \qquad (1)$$

is the nth moment of the weighted line–wave-number distribution [Ref. 1, Eq. (4)]. The weight w_{ab} of a transition is taken as the z part of the strength [$w_{ab} = |(a \mid Z \mid b)|^2$], W is the sum $\sum_{a,b} w_{ab}$, and a and b are the states of the respective configurations in intermediate coupling. For example, μ_1 can also be written

$$\mu_1 = \frac{\sum_{a,b} (a \mid Z \mid b)(b \mid H \mid b)(b \mid Z \mid a) - \sum_{a,b} (b \mid Z \mid a)(a \mid H \mid a)(a \mid Z \mid b)}{\sum_{a,b} (a \mid Z \mid b)(b \mid Z \mid a)} , \qquad (2)$$

an expression suitable for the use of the second-quantization formalism.[7] Throughout the following, the results and arguments of Ref. 1, hereafter referred to as I, are largely used.

For the sake of brevity, each single letter l, l', l'', and λ figures in the present paper, either the whole symbol nl, etc., for some atomic subshell, or only the corresponding l orbital quantum number.

II. CONFIGURATIONS WITH THREE OPEN SUBSHELLS

A. Variance of the $\lambda^\nu l^N l'$-$\lambda^\nu l^N l''$ transition array

The array $\lambda^\nu l^N l'$-$\lambda^\nu l^N l''$ is the simple extension of the $nl^N n'l'$-$nl^N n''l''$ array, studied in I, by the addition of one passive subshell λ^ν. The list of products of Slater integrals (PSI) which may occur in the variance σ^2 can be divided into three groups, which are considered successively.

(i) In group 1 are gathered the PSI already occurring in the variances of the arrays $\lambda^\nu l'$-$\lambda^\nu l''$ or $l^N l'$-$l^N l''$, where they can be calculated with Table III of I. In the former case, for example, choosing the couplings of the states as $a=[(\lambda^\nu l')\alpha_1 J_1, l^N \alpha_2 J_2]JM$ and $b=[(\lambda^\nu l'')\alpha_1' J_1', l^N \alpha_2 J_2]J'M$ is convenient for the argument. In the calculation of σ^2, the considered part of the energy does not depend on $\alpha_2 J_2$. The latter quantum numbers only appear in the matrix elements of the transition operator Z, through the same sum in the numerator as in the denominator of μ_2 [Eq. (1)]. Therefore, that part of μ_2 does not depend on N, and the contributions to σ^2 of the PSI of group 1 are simply the applications of Table III of I to both $\lambda^\nu l'$-$\lambda^\nu l''$ and $l^N l'$-$l^N l''$.

(ii) In group 2 are gathered the PSI where λ and/or l occur only once. The product $F^k(\lambda,l')G^{k'}(l,l'')$ can be taken as an example. In this case, it is first deduced that the second-quantization operator Op [Eq. (9) in I] for the second-order moment μ_2 contains only one creation-annihilation pair of operators for λ electrons. Then, the corresponding part of σ^2 depends on the number ν through a binomial factor of the form $\alpha\nu+\beta$. But this binomial evidently vanishes when $\nu=0$ and $4\lambda+2$; therefore, it is zero for any value of ν, and it can be concluded that the PSI of group 2 do not contribute to σ^2.

(iii) In group 3 are gathered the remaining PSI where only λ and l occur. Here the adopted couplings are $[(\lambda^\nu l^N)\alpha_1 J_1, l'j']JM$ and $[(\lambda^\nu l^N)\alpha_1 J_1, l''j'']J'M$, and the sums over J, M, J', j', and j'' reduce to a multiple of $(2J_1+1)$, i.e., to the statistical weight of the relevant level of $\lambda^\nu l^N$ (a similar property has been exploited in I, Sec. III B, for explaining why expressions D_1 and E_1 are almost identical). Therefore, Table I of I, which gives the variance σ^2 of the distribution of the level energies with $(2J+1)$ as statistical weight, is useful. However, it is evident that, in the case where any Slater integral containing one l' or l'' electron is zero and where the other integrals have identical values in both configurations, σ^2 is zero because all the lines have the same energy. In this way, it can be shown that the contributions to σ^2 (of the transition array) of the PSI of group 3 are simply the application of Table I of I to $\lambda^\nu l^N$, each Slater integral in this Table being replaced by its increment from configuration A to configuration B. For example, the formula for the quantity D_7 then contains the product

$$[F_B^k(\lambda l)-F_A^k(\lambda l)][G_B^{k'}(\lambda l)-G_A^{k'}(\lambda l)] .$$

At this point, it should also be indicated that in the quoted formula for D_7 in Table I of I a misprint occurred in the $6j$ symbol, which should evidently read

$$\begin{Bmatrix} l' & l' & k \\ l & l & k' \end{Bmatrix}$$

as in formula E_4 of Table III of the same article. The contributions to σ^2 coming from the spin-orbit integrals are easier to compute. The result is

$$E_8+\frac{\lambda(\lambda+1)\nu(4\lambda-\nu+2)(\Delta\zeta_\lambda)^2}{4(4\lambda+1)} , \tag{3}$$

where E_8 is given in Table III of I and where the second term makes the contributions of the λ and l electrons formally identical.

In the upper part of Table I, the expansion of the variance σ^2 of the array $ss's''$-$ss'p$ is a simple example of application of the present results.

B. Variance of the $\lambda^\nu l^{N+1}$-$\lambda^\nu l^N l'$ transition array

The array $\lambda^\nu l^{N+1}$-$\lambda^\nu l^N l'$ is the extension of the nl^{N+1}-$nl^N n'l'$ array, studied in I, by the addition of one passive subshell λ^ν. It is convenient to define five groups of PSI, among which the first three resemble those of Sec. II A.

(i) In group 1 are gathered the PSI already occurring in the variance of the array l^{N+1}-$l^N l'$, they can be calculated with Table II of I, a conclusion which is reached in the same way as for group 1 in Sec. II A.

(ii) In group 2 are gathered the PSI where λ occurs only once. The same argument as for group 2 in Sec. II A shows that they do not contribute to σ^2.

(iii) In group 3 are gathered the PSI where only λ appears, i.e., where it appears four times. The argument is analogous to that for group 3 in Sec. II A. The contributions to σ^2 of the PSI of group 3 are simply the application to λ^ν of formula D_1 in Table I of I, each Slater integral being replaced by

TABLE I. Examples of variance expressions for transition arrays A-B. For the array sd^2-sdp, only the PSI of group 5 in Sec. II B are listed.

Array $ss's''$-$ss'p$:

$$\sigma^2=3\{[G^0(s,s'')]^2+[G^0(s',s'')]^2\}/4-[G^0(s,s'')G^1(s,p)+G^0(s',s'')G^1(s',p)]/2$$
$$+\{[G^1(s,p)]^2+[G^1(s',p)]^2\}/12+3[G_A^0(s,s')-G_B^0(s,s')]^2/4+\zeta_p^2/2$$

Array sd^2-sdp:

$$(\text{part of }\sigma^2)=4[G_A^2(s,d)]^2/75-4G_A^2(s,d)G_B^2(s,d)/75+3[G_B^2(s,d)]^2/100$$
$$-4G_A^2(s,d)G^1(s,p)/45-G_B^2(s,d)G^1(s,p)/90+[G^1(s,p)]^2/12$$

its increment from configuration A to configuration B.

(iv) Group 4 contains the PSI where λ appears either three times, or only twice, provided that these two λ's occur in the same integral, $F^k(\lambda,\lambda)$ (in the former case, the latter integral necessarily occurs). The contributions to σ^2 depend on v through a polynomial of order 3 and 2, respectively, and they vanish for $v=0$, 1, $(4\lambda+1)$, and $(4\lambda+2)$, because of the occurence of an $F^k(\lambda,\lambda)$ integral. Therefore, the PSI of group 4 do not contribute to σ^2.

(v) Group 5 contains the PSI where λ occurs once in each integral. These PSI are formed from the Slater integrals of the three types (λ,l) (in configuration A or B) and (λ,l'). The v dependence of their contributions to σ^2 is a polynomial of order 2 which vanishes for $v=0$ and $(4\lambda+2)$, namely $v(4\lambda-v+2)$. The N dependence can be found in the same way, i.e, by taking benefit of the particular values of N for which the contributions to σ^2 vanish, except for the PSI with two (λ,l) integrals of the same configuration. For the latter case, other particular examples must be computed explicitly to determine the N dependence completely. The final results are conveniently expressed in terms of the E_i formulas in Table III of I. They are presented in Table II.

The contributions of the spin-orbit integrals to σ^2 can be divided into three types of products: those where no ζ_λ integral occurs (to this type, formula H_7 in Table II of I evidently applies), those where ζ_λ occurs only once (this type brings a null contribution to σ^2, for the same reason as group 2 of PSI in Sec. II A), and those where ζ_λ occurs twice. The latter contribution does not depend on N, so that N can be fixed to zero, and the result is analogous to the first term of E_8 in Table III of I, namely, with proper notations,

$$\frac{\lambda(\lambda+1)v(4\lambda-v+2)(\Delta\zeta_\lambda)^2}{4(4\lambda+1)}\ . \tag{4}$$

TABLE II. Contributions to the variance of the transition array $\lambda^v l^{N+1}$-$\lambda^v l^N l'$ (denoted A-B) of all the products of the Slater integrals in which λ occurs once. Quantities E_i can be found in Table III of I. The notation $l\to\lambda$, for example, means that in these formulas l must be changed into λ.

PSI type	contributions to σ^2
$(\lambda,l)_A\times(\lambda,l)_A$	$(N+1)(4l-N+1)$
$(\lambda,l)_A\times(\lambda,l)_B$	$-2N(4l-N+1)$
$(\lambda,l)_B\times(\lambda,l)_B$	$N(4l-N+2)$
	$\times\frac{v(4\lambda-v+2)}{4l+1}[E_2+E_3+E_4]$ (with $l\to\lambda$, $l'\to l$, $t\to 1$)
$(\lambda,l)_A\times(\lambda,l')_B$	$(4l-N+1)$
$(\lambda,l)_B\times(\lambda,l')_B$	N
	$\times\frac{v(4\lambda-v+2)}{4l+1}[E_5+E_6+E_7+E_7']$ (with $l\to\lambda$, $l'\to l$, $l''\to l'$, $t\to 1$)
$(\lambda,l')_B\times(\lambda,l')_B$	$v(4\lambda-v+2)[E_2+E_3+E_4]$ (with $l\to\lambda$, $t\to 1$)

In the lower part of Table I, the contribution of the PSI of group 5 [see (v) above] is given for the array sd^2-sdp as a simple example of application of the present results.

C. Variance of the $\lambda^{\nu}l^{N+1}l'^{N'}$-$\lambda^{\nu}l^{N}l'^{N'+1}$ transition array

The array $\lambda^{\nu}l^{N+1}l'^{N'}$-$\lambda^{\nu}l^{N}l'^{N'+1}$ can be considered as the result either of the addition of one passive subshell λ^{ν} to the array $l^{N+1}l'^{N'}$-$l^{N}l'^{N'+1}$ or of one or several l' electrons to the array $\lambda^{\nu}l^{N+1}$-$\lambda^{\nu}l^{N}l'$ considered in Sec. II B. Again, five groups of PSI can be distinguished in σ^2, in complete analogy with those for the latter array. Only three groups contribute, as in Sec. II B.

(i) Group 1, where λ does not appear, can be computed as in $l^{N+1}l'^{N'}$-$l^{N}l'^{N'+1}$. This array was studied by Moszkowski,[5] but under the assumption that any given Slater integral has the same value in both configurations. If this assumption is not made, the formal calculation of the variance σ^2 becomes very cumbersome, so that it was not undertaken in I. However, Moszkowski's assumption corresponds to the cases of frozen-core Hartree-Fock and central-field model potentials, which are currently used,[8,9] so that it may not be necessary here to allow for a variation of the Slater orbitals from one configuration to the other. Consequently, formula (3-7) in Ref. 5 may be used for writing the contribution to σ^2 of the PSI of group 1, namely,

$$\sigma^2(l^{N+1}l'^{N'}\text{-}l^{N}l'^{N'+1}) = \frac{N(4l-N+1)}{4l}\sigma^2(l^2-ll') + \frac{N'(4l'-N'+1)}{4l'}\sigma^2(l'^2-ll') \tag{5}$$

(except for l or $l'=0$, in which case the complementary array $l^{4l-N+1}l'^{4l'-N'+2}$-$l^{4l-N+2}l'^{4l'-N'+1}$ is of a type studied in I).

(ii) Group 3, where only λ appears, contributes exactly as in Sec. II B.

(iii) Group 5, where λ occurs once in each integral, contains 10 types of PSI, six of which are already listed in Table II. In the latter Table, the contributions which appear in the first three lines can be kept unchanged. The results for all 10 PSI are listed in Table III. It can be checked that Table III is altogether invariant when the interchanges $l\leftrightarrow l'$, $N\leftrightarrow N'$, and $A\leftrightarrow B$ are made, and that it reduces to Table II when $N'=0$.

As concerns the contributions of the spin-orbit integrals to σ^2, they can be divided into three types of products. The products where ζ_{λ} occurs once or twice contribute exactly in the same way as in Sec. II B. As for the contributions pertinent to the $l^{N+1}l'^{N'}$-$l^{N}l'^{N'+1}$ array, their dependences on N and N' are very easily determined, and formula H_7 in Table II of I, which corresponds to $N=0$, contains the relevant N- and N'-independent quantities; the expression deduced is

$$\begin{aligned}&\frac{l(l+1)}{4(4l+1)}[(N+1)(4l-N+1)\zeta_{l,A}^2-2N(4l-N+1)\zeta_{l,A}\zeta_{l,B}+N(4l-N+2)\zeta_{l,B}^2]\\&+\frac{l'(l'+1)}{4(4l'+1)}[(N'+1)(4l'-N'+1)\zeta_{l',B}^2-2N'(4l'-N'+1)\zeta_{l',B}\zeta_{l',A}+N'(4l'-N'+2)\zeta_{l',A}^2]\\&-\frac{l(l+1)+l'(l'+1)-2}{4(4l+1)(4l'+1)}[N'(4l-N+1)\zeta_{l,A}\zeta_{l',A}+N(4l'-N'+1)\zeta_{l,B}\zeta_{l',B}\\&\qquad+(4l-N+1)(4l'-N'+1)\zeta_{l,A}\zeta_{l',B}+NN'\zeta_{l',A}\zeta_{l,B}]\,.\end{aligned} \tag{6}$$

This expression is invariant when the interchanges $l\leftrightarrow l'$, $N\leftrightarrow N'$, and $A\leftrightarrow B$ are made, and reduces to formula H_7 of Table II in I when $N'=0$.

D. Average energies (wave numbers) of the transition arrays

Together with the variances of the transition arrays, the mean energies of these arrays can be calculated, as has been done in I for l^{N+1}-$l^{N}l'$ and $l^{N}l'$-$l^{N}l''$. More precisely, the quantity

$$\delta E(C-C') = T_{\mathrm{av}}(C-C') - [E_{\mathrm{av}}(C')-E_{\mathrm{av}}(C)]\,, \tag{7}$$

where $T_{\mathrm{av}}(C-C')$ is the mean energy of the $C-C'$ array (configuration C being lower in energy than configuration C') and $E_{\mathrm{av}}(C)$, the average energy of the configuration C, can be expanded in terms of the energy radial integrals.[10] T_{av} is identical to the moment μ_1 of the weighted line-energy distribution [Eq. (2)].

TABLE III. Contributions to the variance of the transition array $\lambda^{\nu}l^{N+1}l'^{N'}$-$\lambda^{\nu}l^{N}l'^{N'+1}$ (denoted *A-B*) of all the products of the Slater integrals in which λ occurs once. The notations $N\rightarrow 4l-N+1$ and $l\leftrightarrow l'$, for example, mean that N must be changed into $4l-N+1$ and that l and l' must be interchanged.

PSI type	contributions to σ^2
$(\lambda,l)_A\times(\lambda,l)_A$	same as in Table II
$(\lambda,l)_A\times(\lambda,l)_B$	same as in Table II
$(\lambda,l)_B\times(\lambda,l)_B$	same as in Table II
$(\lambda,l)_A\times(\lambda,l')_B$	same as in Table II, multiplied by $(4l'-N'+1)/(4l'+1)$
$(\lambda,l)_B\times(\lambda,l')_B$	same as in Table II, multiplied by $(4l'-N'+1)/(4l'+1)$
$(\lambda,l')_B\times(\lambda,l')_B$	same as in Table II, multiplied by $(N'+1)(4l'-N'+1)/(4l'+1)$
$(\lambda,l)_A\times(\lambda,l')_A$	same as $(\lambda,l)_B\times(\lambda,l')_B$ above, with $l\leftrightarrow l'$, $N\leftrightarrow N'$
$(\lambda,l')_A\times(\lambda,l')_A$	same as $(\lambda,l)_B\times(\lambda,l)_B$ above, with $l\leftrightarrow l'$, $N\leftrightarrow N'$
$(\lambda,l')_A\times(\lambda,l)_B$	same as $(\lambda,l)_A\times(\lambda,l')_B$ above, with $N\rightarrow 4l-N+1$, $N'\rightarrow 4l'-N'+1$
$(\lambda,l')_A\times(\lambda,l')_B$	same as $(\lambda,l)_A\times(\lambda,l)_B$ above, with $l\leftrightarrow l'$, $N\leftrightarrow N'$

The δE quantities can be computed successively for the three arrays studied in Secs. II A–II C.

(i) For the array $\lambda^{\nu}l^{N}l'$-$\lambda^{\nu}l^{N}l''$, three types of integrals must be considered, where, respectively, two λ's, one λ, and no λ's occur. The contributions of the first type of δE clearly do not depend on N; therefore, they are zero, as for $N=0$, which can be found in I. The contributions of the second type depend on ν through some binomial $\alpha\nu+\beta$, as shown in an argument such as the one in Sec. II A (ii); therefore, they are zero, because they vanish for $\nu=0$ and $4\lambda+2$. The contributions of the third type do not depend on ν; therefore, they are zero, as for $\nu=0$, which can be found in I.

(ii) For the array $\lambda^{\nu}l^{N+1}$-$\lambda^{\nu}l^{N}l'$, the same three types of contributions as in (i) appear. The only difference is that for the third type, already present in the case of l^{N+1}-$l^{N}l'$, the contributions are not zero, but are given by Eq. (14) of I.

(iii) For the array $\lambda^{\nu}l^{N+1}l'^{N'}$-$\lambda^{\nu}l^{N}l'^{N'+1}$, the situation is again different only for the contributions to δE which do not depend on λ, namely, those occurring in $l^{N+1}l'^{N'}$-$l^{N}l'^{N'+1}$. For the latter array, it is easy to derive the expression,

$$\delta E(l^{N+1}l'^{N'}-l^{N}l'^{N'+1}) = \frac{4l'-N'+1}{4l'+1}\delta E(l^{N+1}-l^{N}l') - \frac{4l-N+1}{4l+1}\delta E(l'^{N'+1}-l'^{N'}l)\,, \tag{8}$$

which changes in sign when the interchanges $l\leftrightarrow l'$ and $N\leftrightarrow N'$ are made, in agreement with the evident relation $\delta E(C'-C)=-\delta E(C-C')$.

III. GENERALIZATION—CONCLUSION

We deal above with spectra where three open subshells occur, but there exist experimental cases with more open subshells. Neutral uranium is a prototype for atomic spectra with very complex configurations. A long list of its configurations of spectroscopic interest has been computed *ab initio* by Rajnak.[11] In this list, there appear several configurations which contain four open subshells.

In general, an array where one configuration contains m and the other one m or $m-1$ open subshells can be deduced from the three types studied in Sec. II by the addition of $m-3$ passive open subshells. The contributions to σ^2 of all the types of PSI where, at most, three different atomic orbitals occur can be classified in groups, and subsequently computed, exactly as in the corresponding paragraphs of Sec. II. As for the single other type to be considered, the one where four atomic subshells occur once each, its contribution can be shown to be zero through the same kind of argument as for group 2 in Sec. II A. To the quantity δE [Eq. (4)] of any transition array, the $m-3$ passive open subshells quoted above do not contribute.

In conclusion, formulas for the variance and mean energy (wave number) of any electric-dipole transition array between two pure configurations can be found in I or in the present paper. These formulas, although cumbersome, are readily programmable on any computer.

Their numerical applications are relevant provided that:

(i) The effects of configuration mixing can be

neglected, as is often the case in highly ionized spectra;

(ii) the effects of relativity on the radial integrals can be neglected, or accounted for approximately through the use of the nonrelativistic integrals deduced from *ab initio* relativistic integrals[12,13];

(iii) the array of experimental line wave numbers does not split into two or more distinct subarrays, due to some very large spin-orbit effects (for subarray variances, other formulas have been devised and will be published elsewhere);

(iv) the populations of the levels of the upper configuration responsible for the spontaneous-emission "band" under investigation do not deviate too much from a Boltzmann-type distribution.[14]

We are currently applying the present formalism to the interpretation of various plasma spectra obtained from sparks and laser shots.

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