

## Effect of configuration widths on the spectra of local thermodynamic equilibrium plasmas

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We present the extension of the supertransition-array (STA) theory to include configuration widths in the spectra of local thermodynamic equilibrium (LTE) plasmas. Exact analytic expressions for the moments of a STA are given, accounting for the detailed contributions of individual levels within the configurations that belong to a STA. The STA average energy is shifted and an additional term appears in its variance. Various cases are presented, demonstrating the effect of these corrections on the LTE spectrum.

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### I. INTRODUCTION

The supertransition-array (STA) model for calculation of atomic spectra under local thermodynamic equilibrium plasma conditions was presented in Ref. [1] (hereafter denoted as I). The model accounts for *all possible* bound-bound and bound-free radiative transitions. It divides the set of all accessible configurations into subsets, or superconfigurations. Each superconfiguration comprises configurations that are similar in energy. The transition array between two superconfigurations is represented by a Gaussian distribution of spectral strength called a supertransition array. Thus a STA can be considered a collection of unresolved transition arrays (UTAs) [2,3] where, as is usually defined, each UTA represents the set of level-to-level transitions between a pair of configurations.

The first three moments of a STA—total intensity, average energy, and variance—can be calculated exactly using an analytic technique that bypasses the direct summation over all the UTAs it comprises. A convergence procedure is used [4,5] to successively better approximate the detailed structure of the spectral distribution. Each STA is iteratively divided into a number of smaller STAs as described below. The end point of this process, of course, is reached when each STA contains a single UTA. In practice, it is never necessary to carry the procedure past a few iterations to obtain an excellent approximation to the UTA spectrum.

In I, the STA moments were calculated neglecting the energy splitting within configurations, i.e., in the approximation of vanishing UTA widths. In this case each UTA is represented by a single line. Recently, several publications have appeared presenting STA results [4–6] and comparing with experiments [7,8]. These works have in fact been based on an improved model that already includes UTA widths, as well as additional improvements. However, these theoretical developments have not yet been presented.

In this paper we present the STA theory accounting

for UTA widths and shifts. This involves deriving the analytical expressions of the STA moments that explicitly take into account individual level-to-level contributions rather than the configuration averages used in I.

In the next section we reintroduce the concepts and definitions required in the STA model. In Sec. III, we rederive the STA moments, taking into account the individual levels of each superconfiguration, and obtain additional terms representing the energy splitting within configurations. Examples with and without UTA width effects and comparisons with recent experiments are presented in Sec. IV, showing that the UTA widths and shifts significantly affect the spectrum. A discussion and a summary are given in Sec. V.

### II. THE BOUND-BOUND SPECTRUM

The spectral distribution  $S(E)$  of transition strength between bound atomic levels gives the probability per unit length for absorption (or emission) of a photon of energy  $E$  per unit of energy. In the following we will treat absorption: emission is obtained simply by multiplication with the Planck function.

The set of bound-bound transitions may be divided into subsets  $G$  such that

$$S(E) = \sum_G S_G(E), \quad (1)$$

where

$$S_G(E) = \sum_{i,j \in G} N_i w_{ij} P_{ij}(E - E_{ij}). \quad (2)$$

$N_i$  is the density of atoms in the lower level  $i$  and  $w_{ij}$  is the absorption transition probability to level  $j$ ,

$$w_{ij} = \frac{\pi e^2 h}{mc} f_{ij}, \quad (3)$$

where  $f_{ij}$  is the absorption oscillator strength and  $P_{ij}(E - E_{ij})$  is the normalized line shape of the transition, centered at the transition energy  $E_{ij} = E_j - E_i$ .

The total intensity, the average energy, and the variance of the group of transitions  $G$  are given by, respectively,

$$I_G = \int S_G(E) dE = \sum_{i,j \in G} N_i w_{ij} , \quad (4)$$

$$E_G = \frac{\int S_G(E) E dE}{I_G} = \frac{\sum_{i,j \in G} N_i w_{ij} E_{ij}}{I_G} , \quad (5)$$

$$(\Delta E_G)^2 = \frac{\int S_G(E) (E - E_G)^2 dE}{I_G} = \Delta_G^2 + \Delta_P^2 . \quad (6)$$

In Eq. (6),

$$\Delta_G^2 = \frac{\sum_{i,j \in G} N_i w_{ij} (E_{ij} - E_G)^2}{I_G} \quad (7)$$

and

$$\Delta_P^2 \equiv \int P(E - \bar{E})(E - \bar{E})^2 dE \quad (8)$$

is the variance of the single line shape profile assumed equal for all the transitions  $ij$  in  $G$ .

With the moments of  $S_G$  we could present the group of neighboring lines  $G$  as a Gaussian. This, however, has two drawbacks: (i) This approach hides the non-Gaussian profile of the individual line obtained by convolution of the Doppler and collisional profiles and (ii) the variance  $\Delta_P^2$  may be infinite, as is the case when a Lorentzian contribution is important. Instead, we use the moments  $I_G$ ,  $E_G$ , and  $\Delta_G$  to construct the distribution of the line centers in  $G$  as the Gaussian:

$$\Gamma(E - E_G) = \frac{I_G}{\sqrt{2\pi}\Delta_G} \exp \left[ -\frac{1}{2} \left( \frac{E - E_G}{\Delta_G} \right)^2 \right] . \quad (9)$$

In order to describe correctly the contribution of the individual lines to the spectrum we define  $\bar{S}_G$  by convolution of this Gaussian with the individual line shape  $P$  (approximated in our code as a convolution of doppler and Lorentzian collisional profiles, i.e., a Voigt function)

$$\bar{S}_G(E) \equiv \int \Gamma(E' - E_G) P(E - E') dE' . \quad (10)$$

This representation of the spectrum of  $G$  keeps the non-Gaussian contribution of the individual line shape and has the same moments as  $S_G$  of Eq. (1).

Equations (4)–(6) for the moments include detailed summation over all the level-to-level transitions in  $G$ . The detailed transition energies (which were averaged over configurations in  $I$ ) will be retained in the analytic derivation presented in the next section.

So far the theory relates to an unspecified group  $G$  of neighboring lines. In the STA model we are dealing with such well defined groups, i.e., the STAs, having the advantage that their moments can be derived analytically. In the next section we derive exact analytical expressions for the STA moments including energy splitting within configurations.

### III. EVALUATION OF THE SPECTRAL MOMENTS BY SUMMATION OVER LEVELS

In the STA model one such group  $G$  is the collection of transitions originating from a specific superconfiguration by a specified one electron jump. A superconfiguration  $\Xi$  is a collection of ordinary configurations defined symbolically by the product over supershells  $\sigma$ ,

$$\Xi \equiv \prod_{\sigma} \sigma^{Q_{\sigma}} . \quad (11)$$

A supershell, in turn, is the union of energetically adjacent ordinary atomic subshells  $s \equiv j_s \equiv n_s l_s j_s$ . In Eq. (11) the superconfiguration is constructed by distributing the  $Q_{\sigma}$  electrons occupying supershell  $\sigma$  among the subshells in all possible ways subject to  $\sum_{s \in \sigma} q_s = Q_{\sigma}$ :

$$\sigma^{Q_{\sigma}} \equiv \sum_{\sum_{s \in \sigma} q_s = Q_{\sigma}} \prod_s j_s^{q_s} . \quad (12)$$

Clearly, each partition of  $Q_{\sigma}$  in Eq. (11) is an ordinary configuration and the particular one-electron jump from this configuration results in another configuration. The transitions between the two configurations constitute an UTA.

The convergence procedure mentioned above splits supershells to smaller supershells according to their energy spread. For each superconfiguration in its turn, at each step, supershells that give rise to relatively well-separated configurations are preferentially split. The detailed structure of the spectrum is thus gradually revealed, yielding a converging spectrum or any other criteria, such as the Rosseland or the Planck means. The process terminates when a dictated change in these quantities is reached.

The STA model makes the essential approximation that the plasma is hot enough that the Boltzmann factor for the level population does not vary significantly over an ordinary configuration. This approximation is identical to that adopted in the UTA model of Bauche-Arnoult, Bauche, and Klapisch [2,3]. However, this would not be a good assumption for a superconfiguration and the variation of the Boltzmann factor from one configuration to another is accounted for in the STA model.

Another essential point is related to the dependence of the oscillator strength  $f_{ij}$  on the transition energy  $E_{ij}$  between the corresponding two levels. This transition energy can always be written as

$$E_{ij} = E_G + \Delta E_{ij} , \quad (13)$$

where  $\Delta E_{ij}$  is the deviation from the average STA energy. We have found that the dependence of  $f_{ij}$  on  $\Delta E_{ij}$  can be dealt with by modifying the Voigt function of Eq. (10). (However, since the derivation of this modification is nontrivial and its effect is negligible, it is not presented in this paper.) Thus in the calculation of the STA moments,  $f_{ij}$  depends only on  $E_G$ ,

$$f_{ij}(E_{ij}) \approx f_{ij}(E_G) . \quad (14)$$

The explicit dependence of the moments on  $E_{ij}$  is of

course retained.

It can be shown from Eqs. (4)–(7) and (11) that the moments of a STA that contributes to the one-electron jump from orbital  $\alpha$  to orbital  $\beta$  ( $\alpha \Rightarrow \beta$ ,  $\alpha = j_\alpha \equiv n_\alpha l_\alpha j_\alpha \dots$ ) are given in terms of the moments of the UTAs it comprises with no approximation:

$$I_G = \sum_{C \in \Xi} N_C w_C^{(\alpha\beta)}, \quad (15)$$

$$E_G = \frac{\sum_{C \in \Xi} N_C w_C^{(\alpha\beta)} E_C^{(\alpha\beta)}}{I_G}, \quad (16)$$

$$\begin{aligned} \Delta_G^2 &= \sum_{C \in \Xi} \sum_{\substack{i \in C \\ j \in C'}} N_i w_{ij} (E_{ij} - E_C^{(\alpha\beta)} + E_C^{(\alpha\beta)} - E_G)^2 / I_G \\ &= \Delta_G'^2 + \delta[\Delta_G'^2], \end{aligned} \quad (17)$$

with

$$\Delta_G'^2 = \left[ \frac{\sum_{C \in \Xi} N_C w_C^{(\alpha\beta)} (E_C^{(\alpha\beta)})^2}{I_G} - (E_G)^2 \right], \quad (18)$$

$$\delta[\Delta_G'^2] \equiv \frac{\sum_{C \in \Xi} N_C w_C^{(\alpha\beta)} (\Delta_C^{(\alpha\beta)})^2}{I_G}. \quad (19)$$

In Eqs. (15)–(19) the summations are over UTAs represented by the initial and the final configurations  $C$  and  $C'$  connected by the orbital jump  $\alpha \Rightarrow \beta$ . The quantities

$$w_C^{(\alpha\beta)} \equiv \frac{1}{N_C} \sum_{i \in C} N_i \sum_{j \in C'} w_{ij}, \quad N_C = \sum_{i \in C} N_i \quad (20)$$

$$E_C^{(\alpha\beta)} \equiv \frac{\sum_{i \in C} N_i \sum_{j \in C'} w_{ij} E_{ij}}{N_C w_C^{(\alpha\beta)}}, \quad (21)$$

and

$$(\Delta_C^{(\alpha\beta)})^2 \equiv \frac{\sum_{i \in C} N_i \sum_{j \in C'} w_{ij} (E_{ij} - E_C^{(\alpha\beta)})^2}{N_C w_C^{(\alpha\beta)}}, \quad (22)$$

are precisely the first three UTA moments. These were evaluated analytically by Bauche-Arnoult, Bauche, and Klapisch [2,3], assuming statistical distributions  $N_i/N_C = g_i/g_C$ , where  $g_i$  and  $g_C$  are the statistical weights of level  $i$  and configuration  $C$ , respectively. The results were obtained in terms of the *occupation numbers* of  $C$  and radial integrals. A compact presentation of these results is presented, for the case of orthogonal orbitals, in Appendix A. Equation (19) gives the correction to the width of the STA owing to the widths of the UTAs it comprises. The term  $(\Delta_C^{(\alpha\beta)})^2$  was neglected in I. In addition to this correction in the STA width, it is important to note (see I) that the effect of individual lines in Eq. (21) includes the UTA shift of Eq. (A4) and by substitution in Eq. (16) these UTA shifts cause a global shift in the STA average energy.

Since the analytic expressions for the UTA moments do not depend on individual  $i, j$  levels, substituting them into Eqs. (15)–(17) replaces the sum over levels in Eqs. (4)–(6) with sums over configurations (i.e., occupation

numbers). But the number of terms in these summations is, in general, still enormously high. To overcome this difficulty we use the same mathematical techniques developed in I. (i) Using binomial relations we first express the moments as generalized partition functions, which still include the summation *over occupation numbers*. (ii) Recursion formulas are then derived, without approximations, for the generalized partition functions that bypass the need for direct summation over occupation numbers. Details of this procedure are given in Appendix B; the results are summarized below.

### A. Total STA intensity

The total intensity is not affected by the UTA widths and the result is as in I:

$$\begin{aligned} I_G &= b(N/U_{\text{tot}}) X_\alpha g_\alpha g_\beta \left[ \prod_\sigma U_{Q_\sigma}'(g^{\alpha\beta}) \right] \\ &\times \left[ \sum_\kappa (E_G)^{2\kappa-1} \begin{bmatrix} j_\alpha & \kappa & j_\beta \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{bmatrix}^2 (r_{\alpha\beta}^\kappa)^2 \right], \end{aligned} \quad (23)$$

where

$$b = \frac{8\pi^2 e a_0}{3h^4 c^3},$$

$\kappa$  is the rank of the transition multipole (in most cases dipole  $\kappa = 1$  is sufficient), and the statistical weight of shell  $s$  is  $g_s = 2j_s + 1$ . The Boltzmann factor  $X_s$  is

$$X_s = e^{-[(\epsilon_s - \mu)/kT]}, \quad (24)$$

where  $\epsilon_s$  is the energy of orbital  $s$  and  $\mu$  is the chemical potential. The quantity  $r_{\alpha\beta}^\kappa$  is the relativistic radial integral for the electron jump  $\alpha \Rightarrow \beta$ ,

$$Q'_\sigma = Q_\sigma - \delta_{\alpha\sigma}, \quad \delta_{\alpha\sigma} \equiv \begin{cases} 1, & \alpha \in \sigma \\ 0, & \alpha \notin \sigma, \end{cases} \quad (25)$$

$g^{\alpha\beta}$  is the set of modified orbital statistical weights with

$$g_s^{\alpha\beta} = g_s - \delta_{s\alpha} - \delta_{s\beta}, \quad (26)$$

for orbital  $s$  (here  $\delta$  is the Kronecker delta), and

$$U_Q(g) \equiv \sum_{i \in \Xi} g_i e^{(E_i - Q\mu)/kT} \quad (27)$$

is the partition function of the superconfiguration  $\Xi$  occupied by  $Q$  electrons.  $N$  and  $U_{\text{tot}}$  are, respectively, the total number density and the partition function of the system.

### B. Average STA transition energy

The average energy is given by

$$E_G = D_0 + \sum_\sigma \epsilon_{\alpha\beta}^\sigma(D'), \quad (28)$$

where for the supershell  $\sigma$

$$\varepsilon_{\alpha\beta}^{\sigma}(D') = \sum_{n=1}^{Q_{\sigma}} \phi_n^{\sigma}(D') U_{Q_{\sigma}-n}(g^{\alpha\beta}), \quad (29)$$

with

$$\phi_n^{\sigma}(D') \equiv - \sum_{s \in \sigma} g_s^{\alpha\beta} D_s' (-X_s)^n \quad (30)$$

and

$$D_s' = D_s + \left[ \delta_{s\alpha} - \delta_{s\beta} \frac{2j_{\alpha}}{2j_{\beta}} \right] \delta E_{\alpha\beta}. \quad (31)$$

The explicit expressions for the radial one-body and two-body parts  $D_0 = \langle \beta \rangle - \langle \alpha \rangle$ ,  $D_s = \langle s, \beta \rangle - \langle s, \alpha \rangle$ , and for  $\delta E_{\alpha\beta}$  are given in Appendix A.

### C. STA variance

The result for the variance is

$$\Delta_G^2 = \sum_{\sigma=1}^N \Delta_{\alpha\beta}^{\sigma}(D''), \quad (32)$$

$$\Delta_{\alpha\beta}^{\sigma}(D'') = \sum_{n=1}^{Q_{\sigma}} \eta_n^{\sigma}(D'') U_{Q_{\sigma}-n}(g^{\alpha\beta}) / U_{Q_{\sigma}}(g^{\alpha\beta}) - [\varepsilon_{\alpha\beta}^{\sigma}(D')]^2, \quad (33)$$

$$\eta_n^{\sigma}(D'') \equiv \sum_{m=1}^{n-1} \phi_m^{\sigma}(D') \phi_{n-m}^{\sigma}(D') + n \phi_n^{\sigma}(D'^2 + D''^2), \quad (34)$$

where

$$\begin{aligned} (D'^2 + D''^2)_s &\equiv (D'^2)_s + (D''^2)_s, \\ (D'^2)_s &\equiv D_s'^2, \\ (D''^2)_s &\equiv (g_s^{\alpha\beta} - 1) \Delta_s^2, \end{aligned} \quad (35)$$

where  $\Delta_s^2 \equiv (\Delta_s^{\alpha\beta})^2$  was derived in Refs. [2,3]. A compact and convenient presentation for this quantity appears for completeness in Appendix A as well. A comparison with the results of I shows that the UTA widths are included in the STA moments with the replacements  $D_s \rightarrow D_s'$  in the working formulas for the average energy and  $(D_s)^2 \rightarrow (D_s')^2 + (D'')^2$  in the second term of the variance.

The equations above are derived in Appendix B on the basis of the technique developed in I that assumes zeroth-order energies in the Boltzmann factors. This approximation can be removed by a common correction to the STA intensity that converges to the correct first-order results in the convergence procedure. This part is beyond the scope of the present subject and will be reported elsewhere.

## IV. EXAMPLES OF THE UTA WIDTH IN PLASMA SPECTRA

The above analytical expressions were incorporated in the STA code and used to calculate the following typical spectra. The first example is the spectrum of the  $2p_{3/2}-3d_{5/2}$  transition array in iron at  $T=59$  eV and ion density  $0.0113$  g/cm<sup>3</sup>, shown in Fig. 1. This transition

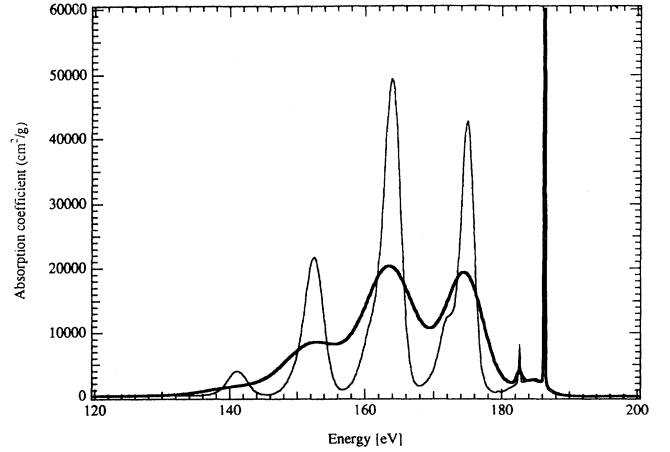


FIG. 1. Absorption spectrum of the  $2p_{3/2}-3d_{5/2}$  transition array in iron at  $T=59$  eV and an ion density of  $0.0113$  g/cm<sup>3</sup>. The heavy and the thin lines describe the spectrum with and without the UTA widths, respectively.

array is the most intense under these conditions. In order to demonstrate the effect of the UTA widths we compare this spectrum with the one obtained without UTA widths as in I. The solid line includes the UTA widths and shifts while the thin line does not. We see in this example that the UTA width smears some of the structure.

In a recent experiment [8] the spectrum of the mixture 0.802 wt. % Fe+0.198 wt. % NaF was obtained at  $T=59$  eV and an ion density of  $0.0113$  g/cm<sup>3</sup> (the same plasma conditions were used in Fig. 1). In Fig. 2 we show the calculated and the experimental transmission spectra. In this calculation, in addition to the bound-bound transitions we have also included the bound-free and free-free transitions as well as scattering. The main features are

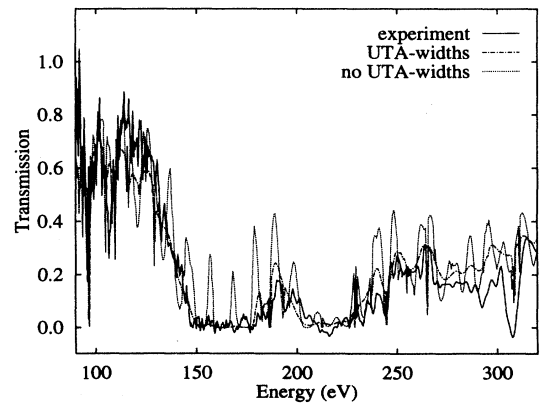


FIG. 2. Total transmission spectrum of the mixture 0.802 wt. % Fe+0.198 wt. % NaF at  $T=59$  eV and an ion density of  $0.0113$  g/cm<sup>3</sup> (the same plasma conditions were used in Fig. 1). The dashed and the dotted lines describe the transmission with and without the UTA widths, respectively. The solid line is the experimental result.

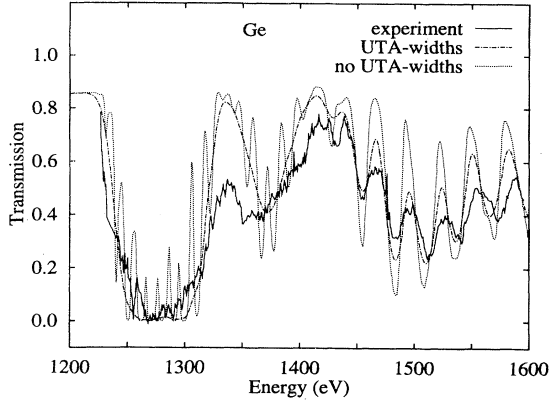


FIG. 3. Total transmission spectrum of Ge at  $T=68$  eV and an ion density of  $0.05$  g/cm<sup>3</sup>. The dashed and the dotted lines describe the transmission with and without the UTA widths, respectively. The solid line is the experimental result.

dominated by the bound-bound part. The dashed and the dotted lines present the calculations with and without UTA widths, respectively. The effect of UTA widths is obvious here. The third (solid) line in Fig. 2, which is the experimental result of Ref. [8], indicates the importance of UTA widths. In this case the total Rosseland opacity is also measured and within the experimental error agrees with the calculated result of the STA model including UTA widths, whereas the omission of these widths results in a discrepancy of about a factor of 2.

Another experiment reported recently [9] presents the transmission spectrum of Ge assumed at  $T=76$  eV and an ion density of  $0.05$  g/cm<sup>3</sup>. In Fig. 3 we compare the experimental results with calculations for these plasma conditions with and without the inclusion of the UTA widths. The calculated spectra here are for  $T=68$  eV, which fits better to the experiment. Again the effect of the UTA widths is apparent and the agreement between the calculations with UTA widths and the experiment is very satisfactory.

## V. DISCUSSION

In this work we have derived the analytical expressions for the STA moments including the effect of the detailed level structure within the configurations. In this respect the model is therefore equivalent to the detailed term accounting (DTA) approach. However, since it avoids dealing directly with levels, it is much faster. In fact, for heavier atoms where DTA calculations are impossible, the STA model is easily applied. It was shown above that the UTA width does not affect the formal form of the STA moments and with only an alternative definition of the radial quantities this extension, though fairly complicated, is easily incorporated in the numerical code and does not increase the required computer time.

The convergence procedure of the STA method leads in this case to the detailed UTA result where each UTA is a Gaussian with the UTA variance. This assumes that each UTA is completely unresolved. The results of I, on the other hand, ignore the UTA widths and are therefore

equivalent to the detailed configuration accounting approach. The only assumption, still hidden in our corrected model, is that an UTA is completely unresolved. This approximation may overestimate the absorption when the number of lines within an UTA is very small so that the accumulated width of these lines is smaller compared to the UTA width. However, in all the experiments performed so far, as in the examples of Figs. 2 and 3, the effect of the UTA widths on the spectra is very important in reproducing the accurate absorption.

## ACKNOWLEDGMENTS

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## APPENDIX A: COMPACT FORM FOR THE ANALYTICAL EXPRESSIONS OF THE UTA MOMENTS

The results obtained by Bauche-Arnoult, Bauche, and Klapisch [2,3] for the UTA moments can be rewritten in a concise form. We present here the results for the case where the same set of orbitals is used for the initial and the final configurations. Furthermore, we extend the formulas to transitions of the more general case with electric multipoles of rank  $\kappa$  (not necessarily dipole). The working formulas are obtained as follows.

### 1. The UTA average energy

The UTA average energy

$$E_C^{(\alpha\beta)} = E_C'^{(\alpha\beta)} + \delta E_C'^{(\alpha\beta)} \quad (\text{A1})$$

includes a shift  $\delta E_C'^{(\alpha\beta)}$  from the difference between the corresponding configuration energy averages (first order),

$$E_C'^{(\alpha\beta)} \equiv E_{C'} - E_C, \quad (\text{A2})$$

$$E_C'^{(\alpha\beta)} = D_0 + \sum_s (q_s - \delta_{\alpha\alpha}) D_s. \quad (\text{A3})$$

The shift is given by

$$\delta E_C'^{(\alpha\beta)} = \left[ \frac{q_\alpha - 1}{2j_\alpha} - \frac{q_\beta}{2j_\beta} \right] 2j_\alpha \delta E(j_\alpha^2 \rightarrow j_\alpha j_\beta). \quad (\text{A4})$$

The radial quantities in (A3) and (A4) are

$$D_0 \equiv D_0^{j_\alpha j_\beta} = \langle j_\beta \rangle - \langle j_\alpha \rangle, \quad (\text{A5})$$

$$\langle j_s \rangle \equiv \langle j_s | h_D | j_s \rangle, \quad (\text{A6})$$

$$D_s^{j_\alpha j_\beta} \equiv \langle j_s, j_\beta \rangle - \langle j_s, j_\alpha \rangle, \quad (\text{A7})$$

$$\begin{aligned} \langle j_r, j_s \rangle = & F^0(j_r, j_s) - \frac{g_{j_s}}{(g_{j_s} - \delta_{j_r, j_s})} \\ & \times \sum_k \begin{bmatrix} j_r & k & j_s \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{bmatrix}^2 G^k(j_r, j_s), \end{aligned} \quad (\text{A8})$$

where  $h_D$  is the Dirac single-particle zeroth-order Hamiltonian,  $F^k$  and  $G^k$  are the Slater integrals, and

$$2j_\alpha \delta E(j_\alpha^2 \rightarrow j_\alpha j_\beta) = F^{j_\alpha j_\beta} + G^{j_\alpha j_\beta}, \quad (\text{A9})$$

with

$$F^{j_\alpha j_\beta} \equiv - \sum_{k \neq 0 \text{ even}} g_{j_\alpha} g_{j_\beta} \begin{Bmatrix} k & j_\alpha & j_\alpha \\ \kappa & j_\beta & j_\beta \end{Bmatrix} \begin{Bmatrix} j_\alpha & k & j_\alpha \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{Bmatrix} \\ \times \begin{Bmatrix} j_\beta & k & j_\beta \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{Bmatrix} F^k(j_\alpha j_\beta), \\ G^{j_\alpha j_\beta} = \sum_k \frac{g_{j_\alpha} g_{j_\beta} \delta_{k,\kappa} - (2\kappa + 1)}{2\kappa + 1} \begin{Bmatrix} j_\alpha & k & j_\beta \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{Bmatrix}^2 G^k(j_\alpha j_\beta) \quad (\text{A10})$$

depending on the radial orbitals, but not on the shell occupation numbers.

We thus notice immediately from (A4) that the UTA shift has the same dependence on the occupation numbers as in (A3), i.e.,

$$E_C^{(\alpha\beta)} = D_0 + \sum_s (q_s - \delta_{s\alpha}) D'_s, \quad (\text{A11})$$

with the following substitution:

$$D'_s = D_s + \left[ \delta_{s\alpha} - \delta_{s\beta} \frac{2j_\alpha}{2j_\beta} \right] \delta E_{\alpha\beta}. \quad (\text{A12})$$

## 2. The UTA variance

The compact form of the results of Bauche-Arnoult, Bauche, and Klapisch [2,3] for the UTA variance is

$$(\Delta_C^{(\alpha\beta)})^2 = \sum_s (q_s - \delta_{s\alpha})(g_s - q_s - \delta_{s\beta}) \Delta_s^2, \quad (\text{A13})$$

where  $\Delta_s^2$  depends only on radial integrals but not on occupation numbers. This general form of the occupation numbers dependence as written in Eq. (A13) is not apparent in Refs. [2,3]. The explicit expressions for  $\Delta_s^2$  of (A13) is

$$\Delta_s^2 = \frac{\Delta^2(j_s j_\alpha j_s j_\beta)}{2j_s - \delta_{s\alpha} - \delta_{s\beta}}, \quad (\text{A14})$$

where  $\Delta^2(j_s j_\alpha j_s j_\beta)$  for  $s = \alpha, \beta$  and  $s \neq \alpha, \beta$  are given below for simplicity in terms of the factored radial Slater integrals

$$\bar{F}^k(j_a j_b) \equiv X^k(j_a j_b, j_a j_b), \\ \bar{G}^k(j_a j_b) \equiv X^k(j_a j_b, j_b j_a), \quad (\text{A15})$$

where

$$X^k(j_a j_b, j_c j_d) \equiv \langle j_a \| C^{(k)} \| j_c \rangle \langle j_b \| C^{(k)} \| j_d \rangle \\ \times R^k(j_a j_b, j_c j_d), \quad (\text{A16})$$

$$\langle j \| C^{(k)} \| j' \rangle = (-1)^{j+1/2} [j, j']^{1/2} \begin{Bmatrix} j & k & j' \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{Bmatrix} \Pi(lkl'), \quad (\text{A17})$$

and  $\Pi(lkl') = 1$  when 1,  $k$ , and  $l'$  obey the triangle condition and  $l + l' + k$  is even, and zero otherwise:

$$\Delta^2(j_s j_\alpha j_s j_\beta) = \mathcal{A}_s + \mathcal{B}_s + \mathcal{C}_s + \mathcal{D}_s + \mathcal{E}_s \\ + \mathcal{F}_s(\alpha\beta) + \mathcal{F}_s(\beta\alpha), \quad (\text{A18})$$

where

$$\mathcal{A}_s = \sum_{\nu=\alpha,\beta} \sum_{k \neq 0 \text{ even}} \frac{1}{(2k+1)(2j_s+1)(2j_\nu+1)} \\ \times [\bar{F}^k(j_s, j_\nu)]^2,$$

$$\mathcal{B}_s = \sum_{\nu=\alpha,\beta} \sum_k \sum_{k'} \left[ \frac{\delta_{kk'}}{2k+1} - \frac{1}{(2j_s+1)(2j_\nu+1)} \right] \\ \times \frac{1}{(2j_s+1)(2j_\nu+1)} \\ \times \bar{G}^k(j_s, j_\nu) \bar{G}^{k'}(j_s, j_\nu),$$

$$\mathcal{C}_s = \sum_{\nu=\alpha,\beta} \sum_{k \neq 0} \sum_{k'} 2(-1)^{k'} \begin{Bmatrix} j_\nu & j_s & k' \\ j_s & j_\nu & k \end{Bmatrix} \\ \times \frac{1}{(2j_s+1)(2j_\nu+1)} \\ \times \bar{F}^k(j_s, j_\nu) \bar{G}^{k'}(j_s, j_\nu),$$

$$\mathcal{D}_s = - \sum_k \frac{2}{2k+1} \begin{Bmatrix} j_\alpha & j_\alpha & k \\ j_\beta & j_\beta & \kappa \end{Bmatrix} \frac{1}{(2j_s+1)} \\ \times (-1)^{j_\alpha+j_\beta+1} \bar{F}^k(j_s, j_\alpha) \bar{F}^k(j_s, j_\beta),$$

$$\mathcal{E}_s = 2 \sum_k \sum_{k'} \left[ - \begin{Bmatrix} k & k' & \kappa \\ j_\beta & j_\alpha & j_s \end{Bmatrix}^2 \right. \\ \left. + \frac{1}{(2j_s+1)(2j_\alpha+1)(2j_\beta+1)} \right] \\ \times \frac{1}{(2j_s+1)} (-1)^{j_\alpha+j_\beta+1} \\ \times \bar{G}^k(j_s, j_\alpha) \bar{G}^{k'}(j_s, j_\beta),$$

$$\mathcal{F}_s(\alpha\beta) = 2 \sum_{k \neq 0 \text{ even}} \sum_{k'} \left[ -(-1)^{k'} \begin{Bmatrix} j_\beta & j_\beta & k \\ j_\alpha & j_\alpha & \kappa \end{Bmatrix} \right. \\ \left. \times \begin{Bmatrix} j_\beta & j_\beta & k \\ j_s & j_s & k' \end{Bmatrix} \right] \\ \times \frac{1}{(2j_s+1)} (-1)^{j_\alpha+j_\beta+1} \\ \times \bar{F}^k(j_s, j_\alpha) \bar{G}^{k'}(j_s, j_\beta),$$

and  $\mathcal{F}(\beta\alpha)$  is the same as  $\mathcal{F}(\alpha\beta)$  with the interchange  $\alpha \leftrightarrow \beta$ . For  $s = \alpha$ ,

$$\Delta^2(\mathbf{j}_s, \mathbf{j}_\alpha, \mathbf{j}_s, \mathbf{j}_\beta) = \mathcal{A}'_s + \mathcal{B}'_s + \mathcal{C}'_s + \mathcal{D}'_s + \mathcal{E}'_s + \mathcal{F}'_s, \quad (A19)$$

$$\mathcal{A}'_s = \sum_{k \neq 0} \sum_{\text{even } k'} \sum_{\text{even } k'} \left[ \frac{\delta_{kk'}}{2k+1} - \frac{1}{2j_\alpha(2j_\alpha+1)} + \begin{Bmatrix} j_\alpha & j_\alpha & k \\ j_\alpha & j_\alpha & k' \end{Bmatrix} \right] \frac{1}{2j_\alpha(2j_\alpha+1)} \bar{F}^k(\mathbf{j}_\alpha, \mathbf{j}_\alpha) \bar{F}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\alpha),$$

$$\mathcal{B}'_s = \sum_{k \neq 0} \sum_{\text{even } k'} \sum_{\text{even } k'} \begin{Bmatrix} j_\alpha & j_\alpha & k' \\ j_\beta & j_\beta & \kappa \end{Bmatrix} \left[ -\frac{\delta_{kk'}}{2k+1} - \begin{Bmatrix} j_\alpha & j_\alpha & k' \\ j_\alpha & j_\alpha & k \end{Bmatrix} + \frac{1}{2j_\alpha(2j_\alpha+1)} \right] \\ \times \frac{1}{j_\alpha} (-1)^{j_\alpha+j_\beta+1} \bar{F}^k(\mathbf{j}_\alpha, \mathbf{j}_\alpha) \bar{F}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\beta),$$

$$\mathcal{C}'_s = \sum_{k \neq 0} \sum_{\text{even } k'} \left[ -\begin{Bmatrix} k & k' & \kappa \\ j_\beta & j_\alpha & j_\alpha \end{Bmatrix}^2 + \begin{Bmatrix} j_\beta & j_\beta & k \\ j_\alpha & j_\alpha & \kappa \end{Bmatrix} \begin{Bmatrix} j_\beta & j_\beta & k \\ j_\alpha & j_\alpha & k' \end{Bmatrix} - \frac{1}{2j_\alpha(2j_\alpha+1)} \left[ \frac{\delta_{k'\kappa}}{2\kappa+1} - \frac{1}{2j_\beta+1} \right] \right] \\ \times \frac{1}{j_\alpha} (-1)^{j_\alpha+j_\beta+1} \bar{F}^k(\mathbf{j}_\alpha, \mathbf{j}_\alpha) \bar{G}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\beta),$$

$$\mathcal{D}'_s = \sum_{k \neq 0} \sum_{\text{even } k'} \sum_{\text{even } k'} \left[ \frac{\delta_{kk'}}{(2k+1)(2j_\beta+1)} - \begin{Bmatrix} j_\alpha & j_\alpha & k \\ j_\alpha & \kappa & j_\beta \\ k' & j_\beta & j_\beta \end{Bmatrix} - \frac{1}{2j_\alpha} \begin{Bmatrix} j_\alpha & j_\beta & \kappa \\ j_\beta & j_\alpha & k \end{Bmatrix} \begin{Bmatrix} j_\alpha & j_\beta & \kappa \\ j_\beta & j_\alpha & k' \end{Bmatrix} \right] \frac{1}{2j_\alpha} \bar{F}^k(\mathbf{j}_\alpha, \mathbf{j}_\beta) \bar{F}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\beta),$$

$$\mathcal{E}'_s = \sum_k \sum_{k'} \left[ \frac{\delta_{kk'}}{(2k+1)(2j_\beta+1)} - \begin{Bmatrix} j_\alpha & j_\beta & k \\ j_\beta & \kappa & j_\alpha \\ k' & j_\alpha & j_\beta \end{Bmatrix} - \frac{1}{2j_\alpha} \left[ \frac{\delta_{k\kappa}}{2\kappa+1} - \frac{1}{(2j_\beta+1)} \right] \left[ \frac{\delta_{k'\kappa}}{2\kappa+1} - \frac{1}{(2j_\beta+1)} \right] \right] \\ \times \frac{1}{2j_\alpha} \bar{G}^k(\mathbf{j}_\alpha, \mathbf{j}_\beta) \bar{G}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\beta),$$

$$\mathcal{F}'_s = \sum_{k \neq 0} \sum_{\text{even } k'} \left[ \frac{-1}{2j_\beta+1} \begin{Bmatrix} j_\beta & j_\beta & k \\ j_\alpha & j_\alpha & k' \end{Bmatrix} - (-1)^{j_\alpha+j_\beta} \begin{Bmatrix} k & k' & \kappa \\ j_\alpha & j_\beta & j_\beta \end{Bmatrix} \begin{Bmatrix} k & k' & \kappa \\ j_\beta & j_\alpha & j_\alpha \end{Bmatrix} + \frac{1}{2j_\alpha} \begin{Bmatrix} j_\alpha & j_\beta & \kappa \\ j_\beta & j_\alpha & k \end{Bmatrix} \left[ \frac{\delta_{k'\kappa}}{2\kappa+1} - \frac{1}{2j_\beta+1} \right] \right] \frac{1}{j_\alpha} \bar{F}^k(\mathbf{j}_\alpha, \mathbf{j}_\beta) \bar{G}^{k'}(\mathbf{j}_\alpha, \mathbf{j}_\beta).$$

#### APPENDIX B: STA MOMENTS INCLUDING UTA WIDTHS

Equation (13) for the STA average energy reads

$$E_G = \frac{\sum_C N_C w_C^{(\alpha\beta)} E_C^{(\alpha\beta)}}{I_G}. \quad (B1)$$

It is seen immediately that the shifted and the unshifted average energies (A3) and (A11), respectively, have exactly the same occupation number dependence. Since the derivation procedure of I operates only on occupation numbers, the result for the STA average energy has the same form as that of I:

$$E_G = D_0 + \sum_\sigma \varepsilon_{\alpha\beta}^\sigma(D), \quad (B2)$$

$$\varepsilon_{\alpha\beta}^\sigma(D) = \sum_{n=1}^{Q_\sigma} \phi_n^\sigma(D) U_{Q_\sigma-n}(g^{\alpha\beta}) / U_{Q_\sigma}(g^{\alpha\beta}), \quad (B3)$$

with the substitution  $D_s \rightarrow D'_s$  of (A12). The final results appear in Eqs. (28)–(31).

The derivation for the STA variance due to configuration widths is more complex. As shown in Eq. (17), the correction of the STA variance due to UTA variances is given by

$$\delta[\Delta_G^2] = \frac{\sum_{C \in \mathcal{E}} N_C w_C^{(\alpha\beta)} (\Delta_C^{(\alpha\beta)})^2}{I_G}. \quad (B4)$$

Using the relations [1]

$$N_C = \frac{N}{U} \prod_{s \in C} \begin{pmatrix} g_s \\ q_s \end{pmatrix} X_s^{q_s}, \quad (\text{B5})$$

$$w_C^{(\alpha\beta)} = b(E_G^{(\alpha\beta)})^{2\kappa-1} q_\alpha (g_\beta - q_\beta) \times \begin{pmatrix} j_\alpha & \kappa & j_\beta \\ \frac{1}{2} & 0 & -\frac{1}{2} \end{pmatrix} (r_{\alpha\beta}^{(\kappa)})^2, \quad (\text{B6})$$

$$I_G = \sum_{C, C' \in G} N_C w_C^{(\alpha\beta)} \quad (\text{B7})$$

together with (A13) gives, by substitution in (B2),

$$\delta[\Delta_G^2] = \frac{\sum_C \sum_r \Delta_r^2 (q_r - \delta_{r\alpha})(g_r - \delta_{r\beta} - q_r) q_\alpha (g_\beta - q_\beta) \prod_{s \in C} \begin{pmatrix} g_s \\ q_s \end{pmatrix} X_s^{q_s}}{\sum_C q_\alpha (g_\beta - q_\beta) \prod_{s \in C} \begin{pmatrix} g_s \\ q_s \end{pmatrix} X_s^{q_s}}. \quad (\text{B8})$$

It should be mentioned that the  $3j$  symbol in Eq. (B6) contains the hidden triangular and the parity conditions  $\Delta(l_\alpha, \kappa, l_\beta)$  and  $l_\alpha + \kappa + l_\beta$  even. Using the binomial relations

$$q \begin{pmatrix} g \\ q \end{pmatrix} = g \begin{pmatrix} g-1 \\ q-1 \end{pmatrix}, \quad (g-q) \begin{pmatrix} g \\ q \end{pmatrix} = g \begin{pmatrix} g-1 \\ q \end{pmatrix} \quad (\text{B9})$$

we can follow the steps below:

$$\begin{aligned} \delta[\Delta_G^2] &= \frac{\sum_{C \in \Xi} \sum_r \Delta_r^2 q_r^\alpha (g_r^\beta - q_r) \prod_s \begin{pmatrix} g_s^{\alpha\beta} \\ q_s^\alpha \end{pmatrix} X_s^{q_s^\alpha}}{\sum_{C \in \Xi} \prod_s \begin{pmatrix} g_s^{\alpha\beta} \\ q_s^\alpha \end{pmatrix} X_s^{q_s^\alpha}} = \frac{\sum_r \Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) X_r \sum_{C \in \Xi} \prod_s \begin{pmatrix} g_s^{\alpha\beta rr} \\ q_s^{\alpha r} \end{pmatrix} X_s^{q_s^{\alpha r}}}{\sum_{C \in \Xi} \prod_s \begin{pmatrix} g_s^{\alpha\beta} \\ q_s^\alpha \end{pmatrix} X_s^{q_s^\alpha}} \\ &= \frac{\sum_r \Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) X_r \prod_{\sigma \in \Xi} \sum_{\sum_{s \in \sigma} q_s = Q_\sigma} \prod_{s \in \sigma} \begin{pmatrix} g_s^{\alpha\beta rr} \\ q_s^{\alpha r} \end{pmatrix} X_s^{q_s^{\alpha r}}}{\prod_{\sigma \in \Xi} \sum_{\sum_{s \in \sigma} q_s = Q_\sigma} \prod_{s \in \sigma} \begin{pmatrix} g_s^{\alpha\beta} \\ q_s^\alpha \end{pmatrix} X_s^{q_s^\alpha}} = \frac{\sum_r \Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) X_r \prod_{\sigma \in \Xi} U_{Q_\sigma^{\alpha r}}(g^{\alpha\beta rr})}{\prod_{\sigma \in \Xi} U_{Q_\sigma^\alpha}(g^{\alpha\beta})}, \quad (\text{B10}) \end{aligned}$$

where, in general,

$$g_s^{abcd\dots} \equiv g_s - \delta_{as} - \delta_{bs} - \delta_{cs} - \delta_{ds} - \dots,$$

$$q_s^{abcd\dots} \equiv q_s - \delta_{as} - \delta_{bs} - \dots,$$

( $g^{abcd\dots}$ ) stands for the set of values for all  $s$ , and

$$Q_\sigma^{ab} \equiv Q_\sigma - \delta_{a\sigma} - \delta_{b\sigma}, \quad \delta_{a\sigma} = \begin{cases} 1, & a \in \sigma \\ 0, & a \notin \sigma. \end{cases} \quad (\text{B11})$$

In Eq. (B10) all the terms with the shell  $r$ ,  $r \notin \sigma$ , cancel out from both the nominator and denominator and we obtain

$$\delta[\Delta_G^2] = \sum_r \frac{\Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) X_r U_{Q_{r-1}}(g^{\alpha\beta rr})}{U_{Q_r^\alpha}(g^{\alpha\beta})}, \quad (\text{B12})$$

where

$$Q_r \equiv Q_\sigma - \delta_{\alpha\sigma} \quad (\text{B13})$$

and  $\sigma$  is the supershell containing the shell  $r$ . The appearance of  $r$  twice in the set of statistical weights  $g^{\alpha\beta rr}$  means, according to (B9), that for the  $r$  shell the weight should be reduced by 2 (or 3 in the cases  $r = \alpha, \beta$ ). We

now use the identity [1]

$$U_{Q_\sigma}(g^\gamma) = \sum_{n=0}^{Q_\sigma} (-X_\gamma)^n U_{Q_\sigma - n}(g), \quad (\text{B14})$$

leading to

$$\begin{aligned} U_{Q_{r-1}}(g^{\alpha\beta rr}) &= \sum_{n=0}^{Q_{r-1}} (-X_r)^n U_{Q_{r-1}-n}(g^{\alpha\beta r}) \\ &= \sum_{n=0}^{Q_{r-1}} (-X_r)^n \sum_{m=0}^{Q_{r-1}-n} (-X_r)^m \\ &\quad \times U_{Q_{r-1}-n-m}(g^{\alpha\beta}), \quad (\text{B15}) \end{aligned}$$

and defining  $k = n + m + 1$  we obtain

$$\begin{aligned} U_{Q_{r-1}}(g^{\alpha\beta rr}) &= \sum_{n=0}^{Q_{r-1}} (-X_r)^n \sum_{k=n+1}^{Q_r} (-X_r)^{k-n-1} \\ &\quad \times U_{Q_r-k}(g^{\alpha\beta}). \quad (\text{B16}) \end{aligned}$$

With substitution in (B12)



$$\begin{aligned} \delta[\Delta_G'^2] &= \sum_r \frac{\Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) X_r \sum_{k=1}^{Q_r} k (-X_r)^{k-1} U_{Q_r-k}(g^{\alpha\beta})}{U_{Q_r}(g^{\alpha\beta})} \\ &= \sum_\sigma \frac{-\sum_{k=1}^{Q_\sigma} U_{Q_\sigma-k}(g^{\alpha\beta}) k \sum_{r \in \sigma} \Delta_r^2 g_r^{\alpha\beta} (g_r^{\alpha\beta} - 1) (-X_r)^k}{U_{Q_\sigma}(g^{\alpha\beta})} = \sum_\sigma \frac{\sum_{k=1}^{Q_\sigma} U_{Q_\sigma-k}(g^{\alpha\beta}) k \phi_k^\sigma(\{D''^2\})}{U_{Q_\sigma}(g^{\alpha\beta})}, \end{aligned} \quad (\text{B17})$$

where

$$\phi_n^\sigma(D''^2) \equiv - \sum_{s \in \sigma} g_s^{\alpha\beta} \{D''^2\}_s (-X_s)^n \quad (\text{B18})$$

and

$$\{D''^2\}_s \equiv (g_s^{\alpha\beta} - 1) \Delta_s^2, \quad (\text{B19})$$

the results can be summarized as

$$\delta[\Delta_G'^2] = \sum_{\sigma \in G} \delta[\Delta_\sigma'^2], \quad (\text{B20})$$

where

$$\delta[\Delta_\sigma'^2] = \frac{\sum_{n=1}^{Q_\sigma} n \phi_n^\sigma(D''^2) U_{Q_\sigma-n}(g^{\alpha\beta})}{U_{Q_\sigma}(g^{\alpha\beta})}. \quad (\text{B21})$$

The working formula of I for the variance where the UTA widths were neglected is

$$\Delta_G'^2 = \sum_{\sigma \in G} \Delta_\sigma'^2, \quad (\text{B22})$$

where

$$\begin{aligned} \Delta_\sigma'^{\alpha\beta} &= \Delta_\sigma'^{\alpha\beta}(D) \\ &= \sum_{n=1}^{Q_\sigma} \eta_n^\sigma(D) U_{Q_\sigma-n}(g^{\alpha\beta}) / U_{Q_\sigma}(g^{\alpha\beta}) - (\epsilon_{\alpha\beta}^\sigma(D))^2, \end{aligned} \quad (\text{B23})$$

with

$$\eta_n^\sigma(D) = \sum_{m=1}^{n-1} \phi_m^\sigma(D) \phi_{n-m}^\sigma(D) + n \phi_n^\sigma(D^2). \quad (\text{B24})$$

It is seen immediately that the same working formula holds for the corrected variance with the replacement

$$(D^2)_s \rightarrow (D^2)_s + (D''^2)_s \quad (\text{B25})$$

in the second term of (B24), i.e., the factor in (B23) should be replaced by

$$\eta_n^\sigma(D'') \equiv \sum_{m=1}^{n-1} \phi_m^\sigma(D) \phi_{n-m}^\sigma(D) + n \phi_n^\sigma(D^2 + D''^2), \quad (\text{B26})$$

as presented in the text.

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