Validity of the continuous-slowing-down approximation in electron degradation, with numerical results for argon

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The electron degradation spectrum plays a key role in describing the electron slowing-down processes in matter. The validity of a method to obtain the electron degradation spectrum approximately, i.e., the continuous-slowing-down approximation with full accounting of electron production, is examined. Numerical results derived with this method are compared with the direct solution of the Spencer-Fano equation for argon using the same set of electron-collision cross sections. The method is effective at electron kinetic energies much higher than the first ionization threshold but fails at low electron kinetic energies even when the kinetic energy of an incident electron is high. Reasons for this failure are discussed in detail.

I. INTRODUCTION

After entering matter, an energetic electron loses its kinetic energy through excitation and ionization of atoms or molecules that constitute matter. Energetic secondary electrons join in further excitation and ionization. As a result, various excited species and ions are generated in matter. Evaluation of the yields of such species is important for complete understanding of radiation effects. One method for calculating the yields uses the degradation spectrum[1,2] (or track-length distribution). To obtain the degradation spectrum exactly, we must solve the Spencer-Fano equation (SFE); however, direct numerical solution of the equation is time consuming even on a current computer.[3] In particular, solving the *time-dependent* Spencer-Fano equation[4] is even more time consuming. Therefore it is valuable to explore approximate methods for obtaining the degradation spectrum efficiently.

An approximate treatment of the SFE, the continuous-slowing-down approximation with generation counting (CSDAGC), is easier and more economical. In this method, the contributions of a primary electron, first-generation electrons, second-generation electrons, and so on, are successively calculated and summed. Sato and co-workers[5-10] used the CSDAGC extensively in combination with the binary-encounter cross sections to treat electron degradation in various molecules and mixtures. Green and co-workers[11-13] and Dalgarno, McElroy, and Stewart[14] also used a similar approach.

There have been studies on the validity of the continuous-slowing-down approximation (CSDA). Fano[1] and Spencer and Fano[2] discussed the CSDA, and in the latter article they compared the degradation spectra of a primary electron in the high-energy region from the SFE and the CSDA. Douthat[15] commented on the CSDA. Peterson[16] tested the validity of the CSDA by comparing yields. Klots and Wright[17] compared the degradation spectrum obtained from Monte Carlo calculations with that from the CSDA. Kowari and Sato[18] tested the validity of the CSDAGC by comparing the degradation spectra from the SFE and CSDAGC. Fano and Spencer[19] introduced an index for the appropriateness of the CSDA and discussed the limitations of the CSDA in the *partial* degradation spectrum for the primary electron alone in helium, analyzing the results calculated by Douthat. The index is β as defined by Eq. (17) of Ref. 19. Fano and Spencer showed that β is 1.05 over the whole energy region except near the source energy. It is expected that the value of β, whose difference from unity indicates the degree of departure from the CSDA, is smaller than the ratio of the degradation spectrum of a primary electron from the CSDA to that from the SFE. Soong[20] calculated the degradation spectra in neon from the CSDAGC and the SFE and showed that β can be as large as 1.26 at low energies. Still, it has not been made clear why the CSDAGC fails in the low-energy region.

Electron degradation in argon has been studied recently by using the SFE and a set of comprehensive cross-section data compiled by Eggarter.[21] The present paper uses the same cross-section data and treats the following topics.

(i) A method based on the CSDA taking account of secondary electrons, which is different from the CSDAGC, is derived from the SFE by using the Taylor expansion in the cross-section operator.[4] The method is named the CSDA with full accounting of electron production (CSDAFA) because the degradation spectrum can be calculated without explicitly separating contributions from electrons of successive generations. The CSDAFA and the CSDAGC give the same result.

(ii) Comparisons are made of the degradation spectra in full calculations from the SFE and the CSDAFA and the degradation spectra of a primary electron from both methods.

(iii) An attempt is made to improve the CSDFA by using a simple modification.

II. THEORY

A. Cross sections

Before introducing the SFE, it is necessary to discuss cross sections that enter the equation as coefficients. Let $\sigma_s(T)$ be the cross section for the sth discrete excitation at excitation energy E_s by an electron of energy T. Let $d\sigma_{i\alpha}(T,E)/dE$ be the differential ionization cross section for the αth shell at energy transfer E, where I_α is the threshold. The total ionization cross section for the αth shell is defined as

$$\sigma_{i\alpha}(T)=\int_{I_\alpha}^{(T+I_\alpha)/2}dE\frac{d\sigma_{i\alpha}(T,E)}{dE} \ , \tag{1}$$

and the total inelastic scattering cross section as

$$\sigma_{\text{tot}}(T)=\sum_\alpha \sigma_{i\alpha}(T)+\sum_s \sigma_s(T) \ . \tag{2}$$

The ionization of the αth shell (when it is an inner shell) is followed by the production of an Auger electron of kinetic energy ε_α at the probability η_α.

B. Spencer-Fano equation

The electron degradation spectrum $y(T)$ is a function of electron kinetic energy T; under stationary irradiation is obeys the SFE

$$nK_Ty(T)+U(T)=0 \ , \tag{3}$$

where K_T is the cross-section operator[4] discussed below, n is the number density of atoms in the medium, and $U(T)$ is the energy distribution of source electrons.
The K_T operator is defined as

$$K_Ty(T)=\sum_s \sigma_s(T+E_s)y(T+E_s)+\sum_\alpha \int_{I_\alpha}^{\lambda_\alpha}dE\frac{d\sigma_{i\alpha}(T+E,E)}{dE}y(T+E)$$

$$+\sum_\alpha \int_{2T+I_\alpha}^{T_0}dT'\frac{d\sigma_{i\alpha}(T',T+I_\alpha)}{dE}y(T')+\sum_\alpha \eta_\alpha\delta(T-\varepsilon_\alpha)\int_{I_\alpha}^{T_0}dT'\sigma_{i\alpha}(T')y(T')-\sigma_{\text{tot}}(T)y(T) \ . \tag{4}$$

The symbol λ_α represents the smaller of $T+I_\alpha$ and T_0-T. The sum over the Auger terms obviously runs over inner shells only.

If the incident electron is monoenergetic at T_0, we set $U(T)=\delta(T-T_0)$ and write the corresponding solution $y(T_0,T)$. The ionization yield of the αth shell resulting from a single incident electron with T_0 is calculated as

$$N_{i\alpha}(T_0)=n\int_{I_\alpha}^{T_0}dTy(T_0,T)\sigma_{i\alpha}(T) \ . \tag{5}$$

The yield of $N_s(T_0)$ of the excited state s is calculated similarly; one merely replaces $\sigma_{i\alpha}(T)$ with $\sigma_s(T)$ in Eq. (5).

C. Another expression for the degradation spectrum

The energy distribution of secondary electrons produced with energy T $N(T)$, which corresponds to the third term of the right-hand side of Eq. (4), is given as

$$N(T)=n\sum_\alpha \int_{2T+I_\alpha}^{T_0}dT'\frac{d\sigma_{i\alpha}(T',T+I_\alpha)}{dE}y(T') \ . \tag{6}$$

We introduce $y_1(T)$, the degradation spectrum *of the primary electron only.* This spectrum obeys an equation of the same form as Eq. (3), with K_T replaced by K_T', where

$$K_T'y_1(T)=\sum_s \sigma_s(T+E_s)y_1(T+E_s)$$

$$+\sum_\alpha \int_{I_\alpha}^{\lambda_\alpha}dE\frac{d\sigma_{i\alpha}(T+E,E)}{dE}y_1(T+E)$$

$$-\sigma_{\text{tot}}y_1(T) \ . \tag{7}$$

We write the solution as $y_1(T_0,T)$, corresponding to $y(T_0,T)$, if the incident electron is monoenergetic. Using the energy distribution of secondary electrons $N(T)$ in this particular case, we can express the full degradation spectrum in terms of the degradation spectrum of a primary electron, that is,

$$y(T_0,T)=y_1(T_0,T)+\int N(T')y_1(T',T)dT' \ . \tag{8}$$

The correctness of Eq. (8) is confirmed by operating nK_T' on both sides and recalling the definitions of $y(T_0,T)$, $y_1(T_0,T)$, and $N(T)$.

D. CSDA approach

The first two terms on the right-hand side of Eq. (4) represent the number of electrons that reach energy T as a consequence of energy losses. We suppose that all energy losses are small compared with T and use the Taylor expansion. Neglecting all the terms beyond the first order in energy losses and considering the last term in Eq. (4), we obtain

$$K_T y(T) = \frac{\partial}{\partial T} \left[\left[\sum_s E_s \sigma_s(T) + \sum_\alpha \int_{I_\alpha}^{\mu_\alpha} dE \, E \frac{d\sigma_{i\alpha}(T,E)}{dE} \right] y(T) + \sum_\alpha \int_{2T+I_\alpha}^{T_0} dT' \frac{d\sigma_{i\alpha}(T',T+I_\alpha)}{dE} y(T') \right.$$

$$\left. + \sum_\alpha \eta_\alpha \delta(T-\varepsilon_\alpha) \int_{I_\alpha}^{T_0} dT' \sigma_{i\alpha}(T') y(T') \right] , \tag{9}$$

where μ_α is the smaller of $(T+I_\alpha)/2$ and $T_0 - T$. If we replace μ_α with $(T+I_\alpha)/2$, we obtain the stopping cross section

$$s(T) = \sum_s E_s \sigma_s(T) + \sum_\alpha \int_{I_\alpha}^{(T+I_\alpha)/2} dE \, E \frac{d\sigma_{i\alpha}(T',E)}{dE} . \tag{10}$$

Note that we have kept intact the third and fourth terms, which represent the production of secondary and Auger electrons at energy T. Finally, by substituting Eqs. (9) and (10) into Eq. (3) and integrating over T, we obtain the equation for the CSDA with full accounting of electron production when an incident electron is monoenergetic,

$$y(T) = \left[1 + n \sum_\alpha \int_T^{(T_0-I_\alpha)/2} dT, \int_{2T+I_\alpha}^T dT' \frac{d\sigma_{i\alpha}(T',T+I_\alpha)}{dE} y(T') \right.$$

$$\left. + n \sum_\alpha \eta_\alpha \int_T^{T_0} dT, \delta(T-\varepsilon_\alpha) \int_{I_\alpha}^{T_0} dT' \sigma_{i\alpha}(T') y(T') \right] \Big/ [ns(T)] , \tag{11}$$

where $ns(T)$ is the stopping power.

If we drop in Eq. (11) the term due to the contribution from Auger electrons, the equation amounts to the CSDA with successive generation counting, as used by Sato, Okazaki, and Ohno.[5]

Now, we introduce an *effective* stopping cross section

$$s'(T) = \sum_s E_s \sigma_s(T) + \sum_\alpha \int_{I_\alpha}^{\mu_\alpha} dE \, E \frac{d\sigma_{i\alpha}(T,E)}{dE} , \tag{12}$$

in which the upper limit of the E integration is μ_α rather than $(T+I_\alpha)/2$. Replacing the stopping cross sections in Eq. (11) with the effective stopping cross section, we obtain what we may call the modified CSDA with full accounting of electron production (MCSDAFA).

III. NUMERICAL PROCEDURES

The mesh size used for direct solution of Eq. (3), the SFE, is 1 eV above 20 eV and 0.5 eV below 20 eV. The mesh size for the CSDA is logarithmic in energy T. More specifically, the nth mesh point T_n is taken at $T_0 a^n$, where $a = (0.5)^{1/40} = 0.9828$. The computation necessary to obtain the electron degradation spectrum at incident electron energy 2 keV by using the CSDA is more than ten times faster than that for the direct solution of the SFE. The higher the incident electron energy becomes, the more advantageous is the CSDA.

IV. RESULTS

We treat an argon gas at a pressure of 1 atm and temperature of 0 °C. The thresholds energies for various excitations and ionizations are shown in Table I.

A. Comparison of the SFE and the CSDAFA

Figure 1 shows degradation spectra, calculated from the SFE and the CSDAFA, for an incident electron of

$T_0 = 2$ keV. The solid curves represent the results of the SFE, and the dashed curves those of the CSDAFA. The upper two curves show the full spectra $y(T_0,T)$ and the lower two curves the spectra for the primary electron $y_1(T_0,T)$. Because we have already discussed[3] the detailed structures of the degradation spectrum from the SFE, we now concentrate on the differences between the degradation spectra of the SFE and the CSDAFA.

First, let us compare the results of full calculations. The degradation spectrum calculated from the SFE shows a rapid decrease near the source energy and discrete structures, i.e., the Lewis effect (which are indiscernible on the scale of Fig. 1, however). In contrast, the degradation spectrum from the CSDA does not show a rapid decrease. After the rapid decrease, the degradation spectrum from the SFE becomes lower than that from the CSDA. Both spectra get closer and decrease more slowly as energy decreases. Each spectrum has a minimum at about 300 eV. The degradation spectrum from the SFE has a sharp peak at 200 eV due to Auger

TABLE I. Threshold energies for excitation states and ionization shells.

Designation symbol	Excitation energy (eV)	Optical selection
I	11.79	allowed
II	14.25	allowed
III	14.97	allowed
IV	15.48	allowed
$1S_3 + 1S_5$	12.0	forbidden
$4p$	13.0	forbidden

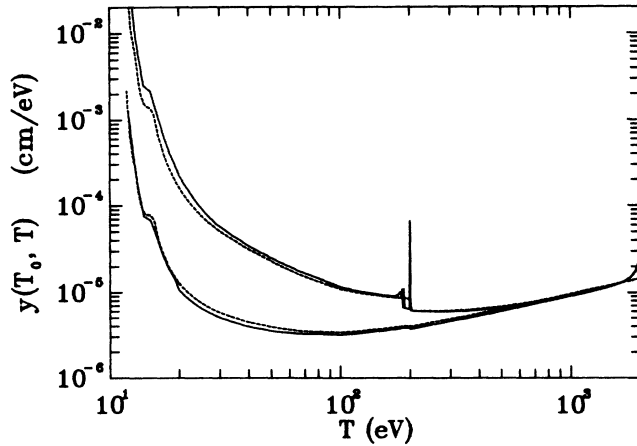| Shell	Ionization energy (eV)
M	15.76
L	250.0

FIG. 1. Electron degradation spectra in Ar at pressure of 1 atm and at a temperature of 0 °C for incident electron energy $T_0 = 2000$ eV, plotted as a function of electron energy T. The solid curves represent the results from the SFE, and the dashed curves those from the CSDAFA. The upper pair of curves show the result of the full degradation spectra, and the lower pair of curves show the contribution by the primary electron only.

electrons from the L-shell ionization and complex structures below the peak (i.e., the Lewis effect due to the L shell). On the other hand, the degradation spectrum from the CSDA increases like a step function at 200 eV. This difference between the two spectra near 200 eV is explained as follows. In the SFE, the degradation spectrum is the ratio of the number of electrons that are generated at energy T due to degradation from higher energies to the total cross section. On the other hand, in the CSDAFA, the degradation spectrum is the ratio of the integrated number of electrons that are generated at every energy above T to the stopping power. Below 200 eV, both spectra increase more rapidly as energy decreases, and the difference between them becomes larger, the degradation spectrum from the SFE being higher than that from the CSDAFA. The general tendency of the differences between the degradation spectra from the SFE and the CSDAFA is the same as that obtained by Kowari and Sato in helium.[18]

Second, we compare $y_1(T_0, T)$, the degradation spectrum of the primary electron only, from the SFE with that from the CSDA. We consider only $y_1(T_0, T)$ below $(T_0 - I_m)/2$, because $y_1(T_0, T)$ is identical with $y(T_0, T)$ between T_0 and $(T_0 - I_m)/2$. The degradation spectrum $y_1(T_0, T)$ from the CSDA is in good agreement with that from the SFE in the whole energy range. Especially in the low-energy region, both spectra agree very well, although the results of full calculations show a large difference. This means that the CSDA gives reasonable $y_1(T_0, T)$ in the whole energy region when incident energy is high. As is well known, the CSDA is justified if the ratio of incident energy to the energy transfer per collision is small, and our results for $y_1(T_0, T)$ illustrate this. Although it is often said that the CSDA becomes invalid because of large energy losses due to occasional but
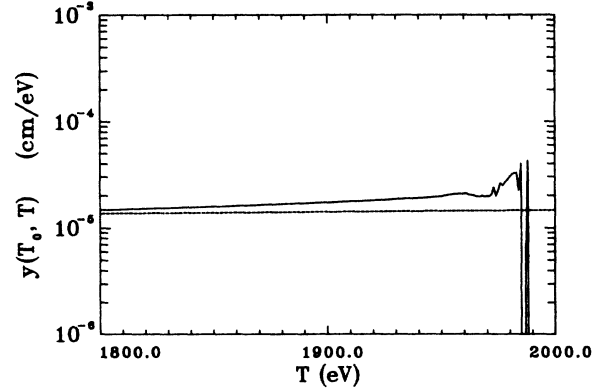

FIG. 2. Electron degradation spectra from the SFE and the CSDAFA near the source energy $T_0 = 2000$ eV. This figure is an enlargement of a part of Fig. 1 near T_0 and compares both spectra.

violent ionization, these large energy losses do not contribute significantly to $y_1(T_0, T)$ (as shown in Fig. 1), and perhaps also to $y(T_0, T)$.

Figure 2 shows an enlargement of the degradation spectra from the SFE and the CSDAFA near the source energy. The degradation spectrum from the SFE (shown in the solid curve) shows oscillations below the source energy; these are the Lewis effect. The degradation spectrum from the CSDAFA (shown in the dashed curve) fails to show the Lewis effect, but approaches the spectrum from the SFE as energy decreases.

B. Comparison of the CSDAFA and the MCSDAFA

Figure 3 shows degradation spectra calculated from the CSDAFA and the MCSDAFA. The degradation spectrum from the MCSDAFA (shown in the long-dash–short-dashed curve) shows a rapid decrease near the source energy. As energy decreases, the MCSDAFA
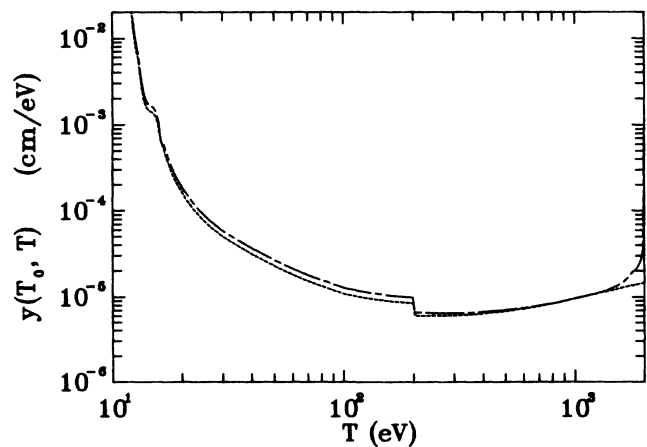

FIG. 3. Electron degradation spectra for $T_0 = 2000$ eV. The dashed curve represents the result from the CSDAFA, and the long-dash–short-dashed curve represents that from the MCSDAFA.

spectrum decreases much more gradually and approaches the degradation spectrum from the CSDAFA (shown in the dashed curve). The degradation spectrum from the MCSDAFA in the transient region where it shows the rapid decrease near the source energy is higher than that from the SFE. As energy further decreases, the degradation spectrum from the MCSDAFA decreases more gradually and approaches that from the CSDAFA. Below 600 eV the MCSDAFA spectrum becomes higher again than the degradation spectrum from the CSDAFA because the number of secondary electrons generated near the source energy is larger than that of the CSDAFA. The degradation spectrum from the MCSDAFA remains higher than that from the CSDAFA; however, below 20 eV it approaches that from the CSADAFA. This means that the MCSDAFA also fails in the low-energy region. The MCSDAFA succeeds in giving high values of $y(T_0, T)$ near the source energy $(1.5 \text{ keV} < T < 2 \text{ keV})$. Nevertheless, it leads to low values of $y(T_0, T)$, comparable to the result of the CSDAFA, at very low energies $(T < 20 \text{ eV})$. The underestimation of the generation of secondary electron ejected with low energy near the source energy from the CSDAFA is not a main reason for the failure of the CSDAFA in the low-energy region.

C. Yields of excitation and ionization

Table II shows the yields of excitation and ionization calculated from the SFE, the CSDAFA, and the MCSDAFA. First, we compare the SFE with the CSDAFA. The yield of the M-shell ionization from the CSDAFA is lower than that from the SFE by 4.8%. Because the difference of the degradation spectra between the SFE and the CSDAFA is large in the low-energy region, the combined yield of the excited states $(1S_3 + 1S_5)$ having cross sections that are appreciable only below 70 eV is expected to show a large difference between the SFE and the CSDAFA. The yield of the excitation from the CSDAFA is indeed lower than that from the SFE by about 30%. This difference is largest among all the excited states treated.

Second, we compare the SFE with the MCSDAFA. The yield of the M-shell ionization from the MCSDAFA is larger than that from the SFE by 13%. This difference

is larger than that between the CSDAFA and the SFE. The overestimation of the M-shell ionization yield is due to the overestimation of the degradation spectrum near the source energy from the MCSDAFA. Because the degradation spectrum from the MCSDAFA is inaccurate in the low-energy region, the yield of the excitation state $(1S_3 + 1S_5)$ from the MCSDAFA is less than that from the SFE by 18%. In the next section, we will discuss the reason why the CSDAFA in full calculations loses its accuracy in the low-energy region.

V. DEGRADATION SPECTRUM IN LOW-ENERGY REGIONS

Figure 4 shows $N(T)$, the energy distribution of secondary electrons that is defined by Eq. (6). The result from the SFE (shown in the solid line) is almost identical with that from the CSDAFA (shown in the dashed line) in the whole energy range except for in the high-energy region. Although the peak at 200 eV due to Auger electrons in the rseult from the SFE is considerably higher than in that from the CSDAFA, the areas under the peaks are consistent within 5% as is understandable from the yields of the L-shell ionization in Table II. Although the CSDAFA is capable of generating $N(T)$ adequately, the CSDAFA loses its validity in the low-energy region as is seen in Fig. 1. The CSDAFA fails in the low-energy region because the CSDAFA does not properly account for the contribution of electrons that are generated with low kinetic energy to the degradation spectrum. This is clear from the well-known fact that the CSDAFA fails in describing the degradation spectrum of an electron incident at low energy, as shown in Refs. 10 and 16.

If we replace $y_1(T', T)$ in Eq. (8) with a reciprocal of the stopping power $ns(T)$, we obtain

$$y(T_0, T) = \frac{1 + \int N(T')dT'}{ns(T)} . \tag{13}$$

Equation (13) is essentially the same as Eq. (11). It is well known that the approximation of $y_1(T', T)$ by $1/ns(T)$,

TABLE II. Comparison of yields of ionization and excitation calculated from the SFE, the CSDAFA, and the MCSDAFA, for incident energy of 2 keV.

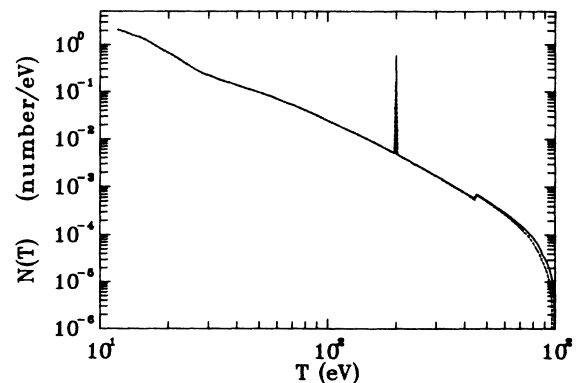| Products	Methods of calculation		
	SFE	CSDAFA	MCSDAFA
M-shell ionization	73.4	69.9	83.1
L-shell ionization	0.573	0.543	0.730
I	13.8	11.2	13.2
II	4.95	4.58	5.42
III	2.75	2.57	3.04
IV	2.88	2.68	3.17
$1S_3 + 1S_5$	2.00	1.43	1.65
$4p$	8.71	6.75	7.82

FIG. 4. Energy distribution of secondary electrons that are ejected with kinetic energy T, defined by Eq. (6). Incident electron energy T_0 is set at 2000 eV. The solid curve represents the results from the SFE, and the dashed curve those from the CSDAFA.

the CSDA, becomes worse as incident electron energy decreases. Because the energy distribution of secondary electrons becomes larger as energy decreases, the replacement of $y_1(T',T)$ by $1/ns(T)$ generates serious errors in the degradation spectrum in the low-energy region. This is seen in Fig. 1.

This circumstance has bearing on some earlier treatments of the SFE. Douthat[15] and Soong[20] attempted to solve the integral form of the SFE by using an iterative method, and they had difficulties in obtaining convergent results in the low-energy region. Incidentally, the integral form of the SFE [Eq. (40) of Ref. 2] is in essence similar to Eq. (13). It is likely that the difficulties may have arisen for a reason similar to the reason that the replacement of $y_1(T',T)$ with $1/ns(T)$ gives a poor result in the low-energy region.

VI. CONCLUSION

The CSDA with full accounting of electron production is derived from the SFE by using the Taylor expansion of the cross section. It is well known that the degradation spectrum from the CSDA (taking account of secondary electrons) loses its validity in the low-energy region, yet the reason for this had not been fully discussed. We have clearly shown the reason.

We have attempted to improve the CSDAFA with the simple modification introduced by the MCSDAFA. The attempt was successful in the sense that the degradation spectrum from the MCSDAFA shows an abrupt decrease near the source energy, as does that from the SFE; however, the degradation spectrum from the MCSDAFA also loses its validity in the low-energy region. We have also identified the reason for difficulties in obtaining convergent results in the low-energy region when the integral form of the SFE is solved by the numerical iteration method.

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[1] U. Fano, Phys. Rev. 92, 328 (1953).

[2] L. V. Spencer and U. Fano, Phys. Rev. 93, 1172 (1954).

[3] K. Kowari, M. Kimura, and M. Inokuti, Phys. Rev. A 39, 5545 (1989).

[4] M. Inokuti, M. Kimura, and M. A. Dillon, Phys. Rev. A 38, 1217 (1988).

[5] S. Sato, K. Okazaki, and S. Ohno, Bull. Chem. Soc. Jpn. 47, 2174 (1974).

[6] K. Okazaki and S. Sato, Bull. Chem. Soc. Jpn. 48, 3523 (1975).

[7] S. Sato, K. Kowari, and K. Okazaki, Bull. Chem. Soc. Jpn. 49, 933 (1976).

[8] K. Okazaki, M. Yamabe, and S. Sato, Bull. Chem. Soc. Jpn. 50, 1409 (1977).

[9] K. Okazaki, E. Oku, and S. Sato, Bull. Chem. Soc. Jpn. 49, 1230 (1976).

[10] K. Kowari and S. Sato, Bull. Chem. Soc. Jpn. 51, 741 (1978).

[11] R. S. Stolarski and A. E. S. Green, J. Geophys. Res. 72, 3967 (1967).

[12] A. E. S. Green and C. A. Barth, J. Geophys. Res. 72, 3975 (1967).

[13] A. E. S. Green, L. R. Peterson, and S. S. Parasad, in Atmospheric Emissions, edited by B. M. McCormac and A. Omholt (Reinhold, New York, 1969), p. 523.

[14] A. Dalgarno, M. B. McElrory, and A. I. Stewart, J. Atmos. Sci. 26, 753 (1969).

[15] D. A. Douthat, Radiat. Res. 61, 1 (1975).

[16] L. R. Peterson, Phys. Rev. 187, 105 (1969).

[17] C. E. Klots and H. Wright, Int. J. Radiat. Phys. Chem. 2, 191 (1970).

[18] K. Kowari and S. Sato, Bull. Chem. Soc. Jpn. 54, 2878 (1981).

[19] U. Fano and L. V. Spencer, Int. J. Radiat. Phys. Chem. 7, 63 (1975).

[20] S. C. Soong, Radiat. Res. 67, 187 (1976).

[21] E. Eggarter, J. Chem. Phys. 62, 833 (1975).