Time dependence of the subexcitation electron distribution generated by high-energy electrons

Michael Dillon and Mineo Kimura

Argonne National Laboratory, Argonne, Illinois 60439 (Received 27 February 1995)

The time-dependent behavior of subexcitation electrons is studied by a technique that employs the *continuous slowing-down approximation*, combined with the *Spencer-Fano equation* via a time-dependent version of the *entry spectrum*. The method is used first to establish generalities about the time behavior of subexcitation electrons generated by initially-high-energy electrons and then is employed to follow the development of subexcitation electron degradation spectra generated by 1-keV electrons in argon.

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I. INTRODUCTION

The interaction of high-energy radiation with matter gives rise to the liberation of energetic electrons that cascade to lower energies by inelastic collisions. The ensuing electron degradation process involves the generation of additional electrons and ions by ionizing collisions, as well as the production of excited states and reactive intermediates by energy transfers, resulting in simple electronic excitation. For energies above the inelastic threshold of atomic and molecular gases, the behavior of the electron energy distribution function, or the electron energy degradation spectrum in pure substances [1-4] and in mixtures [5-7], has been thoroughly discussed in terms of both the steady-state and time-dependent Spencer-Fano equation (SFE). In the final state of degradation in atomic gases, electrons with energies below the first excitation threshold are moderated by momentum transfer alone. Because of their long life and relatively low energies, these electrons are the principal component of innumerable reactions, including ion neutralization; electronic excitation of minor additives; and, in the case of molecular gases, dissociative and nondissociative electron attachment and rotational and vibrational excitation. The importance of electrons with energies below the first electronic excitation threshold was first recognized by Platzmann, who termed them subexcitation electrons [8]. The prime motivation for the present work is the appearance over the past several years of experiments that monitor the products of subexcitation electrons after the introduction of a pulse of electrons in a rare gas [9-11] or a mixture of a rare gas and a minor additive [12-14].

In Sec. II we address the problem from the point of view of an extension of the original SFE. In the subexcitation energy range, the temporal connection is obtained by generating a time-dependent *entry spectrum* from the original numerical solution of the SFE at energies above the excitation threshold. In Sec. II A, a model calculation is used to demonstrate certain common features of subexcitation electron degradation. In Sec. II B, the general method is used to follow the moderation of subexcitation electrons in argon after the introduction of 1-keV electrons.

II. FORMULATION AND RESULTS

A. The time-dependent Spencer-Fano equation

The degradation of energetic electrons in a rare gas occurs in two successive energy domains. The first stage covers the energy range from the initial electron energy T_0 to the threshold of inelastic scattering $E_{\rm th}$. Energy moderation in this range is governed by the time-dependent SFE given by [2,7]

$$v_T^{-1} \frac{\partial z}{\partial t} = nK_T z + u(T, t) , \qquad (1)$$

where T is the electron energy; t is the time; v_T is the electron speed; u is the number of source electrons per unit energy per unit time; n is the gas density; and the incremental degradation spectrum, z (length $\times \text{energy}^{-1} \times \text{time}^{-1}$), is given in terms of the electron density ρ (energy⁻¹), as follows:

$$z = v_T \rho(T, t) . \tag{2}$$

Because all calculations in the present work are density scalable, we have used $n=2.686764\times10^{19}$ molecules/cm³ [at standard temperature and pressure (STP) which is 0°C at 1 atmosphere] as the reference density. In Eq. (1), the cross-section operator K_T is defined in terms of an integral operation, as follows:

$$K_{T}z(T,t) = \sum_{s} \sigma_{s}(T+E_{s})z(T+E_{s},t)$$

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

$$+ \sum_{\alpha} \int_{2T+I_{\alpha}}^{T_{0}} \frac{d\sigma_{i\alpha}(T',T+I_{\alpha})}{dE} z(T',t)dT'$$

$$- \sigma_{tot}(T)z(T,t) + (\text{Auger terms}) . \qquad (3)$$

Here $nK_T z$ is the number of electrons reaching energy T per unit time as a result of all collision processes. In Eq. (3), $d\sigma_{i\alpha}(T,E)/dE$ is the differential ionization cross section for the α th shell at energy transfer E, where the ionization threshold is I_{α} , and σ_s is the cross section for ex-

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citation of the discrete state s with excitation energy E_s . The total ionization cross section from the α th shell is

$$\sigma_{i\alpha}(T) = \int_{I_{\alpha}}^{(T+I_{\alpha})/2} \frac{d\sigma_{i\alpha}(T,E)}{dE} dE , \qquad (4)$$

and the total inelastic cross section is given in terms of $\sigma_{i\alpha}$; and the excitation cross sections σ_s , by

$$\sigma_{\text{tot}}(T) = \sum_{\alpha} \sigma_{i\alpha}(T) + \sum_{s} \sigma_{s}(T) .$$
(5)

Here the sums are taken over s discrete states and α subshells. The partial integro-differential, Eq. (3), is an approximation to the Boltzmann equation known as the cold-gas approximation (CGA) [15,16]. This approximation describes the moderation of electrons in the absence of internal and external fields with energies much above the thermal background.

Certain quantities result from an integration over time. These quantities include the *cumulative degradation spectrum* Z(T,t), defined by

$$Z(T,t) = \int_0^t z(T,\lambda) d\lambda .$$
 (6)

Such quantities are important concepts in radiation physics and chemistry because the time-dependent yield N(t)for a product induced by electron collisions in a gas of density N_k and a cross section σ_k is given by

$$N(t) = N_k \int_{T_1}^{T_2} \sigma_k(T) Z(T, t) dT .$$
 (7)

Integrating Eq. (3) over time shows that Z is the solution of an equation similar to Eq. (1) but with u(T,t) replaced by

$$U(T,t) = \int_0^t u(T,\lambda) d\lambda .$$
(8)

From Eq. (1) it is useful to isolate the quantity $r = K_T z - \sigma_{\text{tot}} z$ -(Auger terms). In an operation analogous to Eqs. (6)–(8), a function R can be defined by

$$R(T,t) = \int_0^t r(T,\lambda) d\lambda . \qquad (9)$$

The quantity $nR(T, \infty)$ has been discussed in considerable detail and is known as the *entry spectrum* [17,18]. Its time-dependent generalization nR(T,t) is essential for understanding the temporal behavior of subexcitation electron yields.

B. Subexcitation electron domain

The second stage of energy degradation, which was the subject of a recent review [19], covers the range from the first electronic threshold at $T_u = E_{\text{th}}$ to some arbitrary lower limit T_L . For the present analysis, T_L should be 10-20 times the thermal energy, so that energy acquired from the medium by the subexcitation electron can be ignored (i.e., the CGA). If m_e is the mass of the electron and M is the mass of the background gas, the mean energy transfer per elastic collision [19] is λT , where

 $\lambda = 2m_e/M$. In the case of an argon medium gas, M = 40and $\lambda = 1.4 \times 10^{-5}$. Hence, the energy step λT is small enough so that the cumulative spectrum Z_s for subexcitation electrons can be obtained by solving the timedependent continuous slowing-down approximation (CSDA). In this limit, the continuity equation [19,20] for electron degradation is

$$v_T^{-1} \frac{\partial Z_s}{\partial t} = n \frac{\partial S(T) Z_s}{\partial T} + nR(T, t) .$$
 (10)

In Eq. (10), the stopping cross section S is given in terms of the momentum transfer cross section σ_m by

$$S(T) = \lambda \sigma_m(T)T . \tag{11}$$

At the steady-state limit $(\partial Z_s / \partial t = 0)$, $R = R(T, \infty)$, and Eq. (10) can be integrated immediately to give

$$Y_{s}(T) = Z_{s}(T, \infty)$$

$$= [S(T)]^{-1} \left[\int_{T}^{T_{u}} R(\lambda, \infty) d\lambda + S(T_{u}) Z(T_{u}, \infty) \right],$$
(12)

where $Z(T_u, \infty)$ is the steady-state solution of Eq. (3) for Z at the lower bound of the first degradation stage.

It is useful to define a CSD time by

$$\tau = \int_{T}^{T_{u}} \frac{d\lambda}{nv_{\lambda}S(\lambda)}$$
(13a)

and the inverse of Eq. (13a) by

$$T = g(\tau) . \tag{13b}$$

Equation (13a) gives the time $\tau(T)$ required for an electron to slow down from T_u to T in a medium gas of density *n* with a stopping cross section S(T). We may then use Eqs. (13a) and (13b) to transform from T to τ in Eq. (10), yielding an equation for the cumulative spectrum Z_s :

$$\frac{\partial SZ_s}{\partial t} = -\frac{\partial SZ_s}{\partial \tau} + nv_T S(g(\tau)) R(g(\tau), t) . \qquad (10')$$

The solution of Eqs. (10) and (10') can be written as the sum of the homogeneous solution, Z_{sh} , and a particular solution, Z_{sp} , as

$$Z_{s}(T,t) = Z_{sh}(T,t) + Z_{sp}(T,t) , \qquad (14a)$$

where

$$Z_{sh}(T,t) = \begin{cases} \frac{S(T_u)}{S(T)} Z(T_u, t-\tau) & \text{for } t \ge \tau \\ 0 & \text{for } t < \tau \end{cases}$$
(14b)

Equation (14b) follows from the solution of $\partial y / \partial t = -\partial y / \partial x$. Here y = F(x - t), and an anticipated boundary requirement is that $Z_s(T_u, t) = Z(T_u, t)$, which is the solution of Eq. (1) at the lower bound of the first degradation stage. The analogous steady-state quantity in the case of Eq. (12) is the second term in the brackets. The particular solution Z_{sp} of Eq. (10') is then given by

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$$Z_{sp}(T,t) = [S(T)]^{-1}n \int_{0}^{t} v_{g(\tau-t+\gamma)} S(g(\tau-t+\gamma)) R(g(\tau-t+\gamma),\gamma) d\gamma$$

$$= [S(T)]^{-1}n \int_{\tau-t}^{\tau} v_{g(\gamma)} S(g(\gamma)) R(g(\gamma),\gamma-\tau+t) d\gamma \quad \text{for } t \leq \tau ,$$

$$Z_{sp}(T,t) = [S(T)]^{-1}n \int_{t-\tau}^{t} v_{g(\tau-t+\gamma)} S(g(\tau-t+\gamma)) R(g(\tau-t+\gamma),\gamma) d\gamma$$

$$= \int_{0}^{t} \int_{0}^{t} V_{g(\tau-t+\gamma)} S(g(\tau-t+\gamma)) R(g(\tau-t+\gamma),\gamma) d\gamma$$

$$= \int_{0}^{t} \int_{0}^{t} V_{g(\tau-t+\gamma)} S(g(\tau-t+\gamma)) R(g(\tau-t+\gamma),\gamma) d\gamma$$

$$= \int_{0}^{t} \int_{0}^{t} V_{g(\tau-t+\gamma)} S(g(\tau-t+\gamma)) R(g(\tau-t+\gamma),\gamma) d\gamma$$

$$= [S(T)]^{-1}n \int_0^\tau v_{g(\gamma)} S(g(\gamma)) R(g(\gamma), t - \tau + \gamma) d\gamma \quad \text{for } t > \tau .$$
(14d)

Examination of Eqs. (14c) and (14d) shows that $Z_{sp}(T_u,t)=0$. As required, the interface condition at $T=T_u$ is then satisfied by $Z_{sh}(T_u,t)=Z(T_u,t)$. In addition, because we will show below that $Z(T_u,\infty)$ is never more than a few percent of $Z_s(T,\infty)$, properties of Z_{sp} will dominate the behavior of Z_s . A characteristic property of the particular solution is readily discerned if we consider the slope of SZ in the SZ- τ plane Taking partial derivatives in Eqs. (14c) and (14d) gives the slope in the two regions as

$$\frac{\partial (SZ)}{\partial t} = \begin{cases} n \int_{0}^{\tau} v_{g(\lambda)} S(g(\lambda)) \partial R(g(\lambda), \lambda - \tau + t) / \partial \tau \, d\lambda + n v_{g(\tau)} S(g(\tau)) R(g(\tau), t) & \text{for } t > \tau , \\ \int_{0}^{\tau} \int_{0}^{\tau} S(g(\lambda)) \partial R(g(\lambda), \lambda - \tau + t) / \partial \tau \, d\lambda + n v_{g(\tau)} S(g(\tau)) R(g(\tau), t) & \text{for } t > \tau , \end{cases}$$
(15a)

$$\frac{\partial \tau}{\partial \tau} = \left[n \int_{\tau-t}^{\tau} v_{g(\lambda)} S(g(\lambda)) \partial R(g(\lambda), \lambda - \tau + t) / \partial \tau \, d\lambda + n v_{g(\tau)} S(g(\tau)) R(g(\tau), t) \right] \text{ for } t \leq \tau .$$
(15b)

These forms show that SZ is continuous at $t = \tau$. In both cases, the second term is positive and the same in each region. However, the first term is negative because $\partial R / \partial t > 0$, from Eq. (9). Moreover, because of the increasing interval of integration in Eq. (15a), the contribution of the first term causes the slope to decrease continuously in the range $0 \le \tau < t$. In the case of Eq. (15b), for $\tau > t$, the interval of integration has a constant value t, while the integration range changes. Hence, from $\tau=0$, $\partial(SZ_{sp})\partial\tau$ decreases until $\tau=t$, where the slope undergoes an alteration in behavior occasioned by the change in the integration limits, as shown in Eqs. (15a) and (15b).

To clarify the foregoing discussion before we cover a practical case of subexcitation electron moderation, we will examine an exactly integrable model that retains physically realistic properties. We can achieve this in Eqs. (10)-(14) by specifying a form for R independent of energy but with a realistic time dependence, such as $nR = [(1-e^{\kappa t}) (eV^{-1})]$. We have chosen $\kappa = 1.6 \times 10^{-8}$ s⁻¹, which is the average reciprocal rise time for nR, computed from the time-dependent entry spectrum employed in the calculation that follows this example. Then as in Refs. [7,19,20], assume that $S = [bT^{1/2} (cm^2 eV)]$. It follows from Eqs. (11) and (13) that $T = g(\tau) = T_u e^{-n\alpha t}$, $\tau = (n\alpha)^{-1} \ln(T_u/T)$, with $b = 2.7230868 \times 10^{-21}$ cm² eV^{1/2} and $\alpha/b = 1.60218925 \times 10^{-12}$ erg eV⁻¹. The magnitude of b, which determines that of σ_m , has been chosen so that results are in rough accordance with those obtained in the numerical calculation that follows. Carrying out the integrations in Eqs. (14c) and (14d) yields

$$\left\{ \alpha T[(n\alpha)^{-1}(e^{n\alpha t}-1) - (n\alpha + \kappa)^{-1}(e^{n\alpha t}-e^{-\kappa t})] \text{ eV}^{-1} \right\} \text{ for } t \le \tau$$
(16a)

$$SZ_{sp}(g(\tau),t) = \begin{cases} \alpha T[(n\alpha)^{-1}(e^{n\alpha\tau}-1) - (n\alpha+\kappa)^{-1}e^{-\kappa t}(e^{(n\alpha+\kappa)\tau}-1)] \text{ eV}^{-1} \end{cases} \text{ for } t > \tau .$$
(16b)

In this case, the steady-state degradation spectrum, Eq. (12), is obtained from Eq. (16b) evaluated at $t = \infty$; $Y = [T^{1/2}(T_u/T-1)/nb \text{ eV}^{-1}]$. The Z_{sp} surface computed from Eqs. (16a) and (16b), displayed in Fig. 1, reveals a ridge in the $SZ(g(\tau), t)$ surface at $t \approx \tau$. As Fig. 1 shows, the ridge is equivalent to a maximum in the SZ- τ plane for every t. Note that the slope in the SZ- τ plane for $\tau > t$,

$$\frac{\partial (SZ_{sp})}{\partial \tau} = \{-n\alpha^2 T[(n\alpha)^{-1}(e^{n\alpha t}-1)-(n\alpha+\kappa)^{-1}(e^{n\alpha t}-e^{-\kappa t})] \text{ eV}^{-1}\} \text{ for } t \leq \tau ,$$

is always negative, while the initial slope

$$\frac{\partial (SZ_{sp})}{\partial \tau} \approx v_{g(\tau)} S(g(\tau)) R(g(\tau), t)$$
$$= [\alpha T(1 - e^{-\kappa t}) \text{ eV}^{-1})] \text{ for } \tau \text{ near } 0$$

is positive. Hence, at each time t, SZ attains a maximum in the SZ- τ plane at some position $\tau \leq t$. In the case of Eqs. (16a) and (16b), the maximum is located in the neighborhood of $\tau \approx t$. In fact, when the rise time of nR, κ^{-1} can be ignored, the maximum is located at exactly $\tau = t$. This may be seen by letting $\kappa \to \infty$ in Eqs. (16a) and (16b), so that $nR \to [\theta(t) \text{ eV}^{-1}]$, where $\theta(t)$ is the step function at t=0. At the limit $\kappa \to \infty$, Eqs. (15a) and (15b) become

$$SZ_{sp}(g(\tau),t) = \begin{cases} [n^{-1}T(e^{n\alpha t}-1) \ (eV^{-1})] \text{ for } t \le \tau \ , \ (17a) \\ [n^{-1}T(e^{n\alpha \tau}-1) \ (eV^{-1})] \text{ for } t > \tau \ . \ (17b) \end{cases}$$

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FIG. 1. Cumulative degradation spectral surface for subexcitation electrons between energies of 0.5 and 12 eV in a model gas at STP. The stopping cross section S(T) of the medium gas is of the form $bT^{1/2}$ and $nR = [(1-e^{-\kappa t}) eV^{-1}]$. The maximum in $SZ_s(g(\tau), t)$ traces a curve in the τ -t plane equivalent to $\tau \approx t$.

The surface described by Eqs. (17a) and (17b) is almost indistinguishable from the plot displayed in Fig. 1, which depicts the $SZ(g(\tau), t)$ surface generated by Eqs. (16a) and (16b). Moreover, the slope in the SZ- τ plane, $\partial (SZ_{sp})/\partial \tau$, from Eq. (17a) is negative, while the same quantity for Eq. (17b) is always positive. Hence, the maximum in the SZ_{sp} surface traces a curve in the τ -t plane that is equivalent to $\tau = t$. Furthermore, because Z_{sh} in Eq. (14) is expected to supply only a minor contribution to the overall Z_s , the global appearance of Z_{sp} in Fig. 1 can be taken as a paradigm for the general behavior of the cumulative yield of subexcitation electrons. In summary, we expect the cumulative yield for subexcitation electrons to possess a maximum in the $Z_s(g(\tau), t)$ - τ plane at $\tau \approx t$ for all t and a corresponding maximum in the $Z_s(T,t)$ -T plane at $T \approx g(t)$.

C. Subexcitation electron degradation in argon

As a test gas we have chosen argon. We use the cross sections complied by Eggarter [21] and the timedependent cumulative yield calculated by Kowari, Inokuti, and Kimura [5]. We combine the two data sources to form the time-dependent entry spectrum,

$$nR(T,t) = n \sum_{s} \sigma_{s}(T+E_{s})Z(T+E_{s},t) + n \sum_{\alpha} \int_{I_{\alpha}}^{T+I_{\alpha}} \frac{d\sigma_{i\alpha}(T+E,E)}{dE} Z(T+E,t)dE + n \sum_{\alpha} \int_{2T+I_{\alpha}}^{T_{0}} \frac{d\sigma_{i\alpha}(T',T+I_{\alpha})}{dE} Z(T',t)dT' = nR_{D}(T,t) + nR_{I1}(T,t) + nR_{I2}(T,t) , \qquad (18)$$

according to Eqs. (3), (6), and (7), where the source terms nR_D , nR_{I1} , and nR_{I2} refer, respectively, to electrons produced by energy lost during excitation of discrete and continuum states and by electron ejection during ionizing collisions.

Figure 2 shows previously calculated [5] values of Z for an electron with 1-keV initial kinetic energy in argon at STP as a function of electron energy and time. Clearly shown in the spectral surface is an Auger spike at $T \cong 200$ eV. Reference to Eq. (18) shows that the influence of these electrons in the subexcitation range is exerted only through Z. The entry spectral surface computed from Eq. (18) is displayed in Fig. 3(d), with individual contributions from nR_D , nR_{I1} , and nR_{I2} shown in Figs. 3(a)-3(c). We note that the ejected-electron component, nR_{12} , contributes about 75% of nR. This observation is expected because ejected electrons in the subexcitation range emanate from ionizing collisions throughout the first stage of degradation, whereas the first two terms in Eq. (18) produce electrons only from initial electrons within a single energy loss of the subexcitation energy range. The sum of the three entry surfaces displayed in Fig. 3(d) reveals a sloped structure with a broad plateau in the neighborhood of 2 eV, caused by the superposition of maxima from nR_D and nR_{I1} .

Before we apply the results of Fig. 3 to the solution of Eqs. (14c) and (14d), we need to compute the homogeneous solution, Eq. (14b), by using the terminal values $Z(T_u,t)$ of the original degradation calculation in stage 1, together with the stopping cross section S shown in Fig. 4. The σ_m values used in Eq. (11) to derive S(T) were obtained by a spline interpolation of a compilation by Hayashi [22], and the quantity $Z(T_u,t)$ used in calculating Eq. (12) was extracted from the Z surface shown in Fig. 2. The subsequent homogeneous Z_{sh} surface is



FIG. 2. Cumulative degradation spectral surface of electrons generated in argon at a constant integrated rate of 1 eV^{-1} , by 1000-eV electrons at STP. The cross sections used in the original calculation were those of Eggarter [21].

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FIG. 3. Integrated rate (in eV^{-1}) of subexcitation electron production or time-dependent *entry spectrum* by 1000-eV electrons in argon at STP. The subexcitation energy range is 0.5-12 eV. The individual integrated rate contributions, defined in Eq. (16), are plotted (a)-(c). A summation of all three contributions is given in (d).



FIG. 4. Stopping cross section for argon according to Eq. (11), plotted as a function of electron energy over a range of energies that includes the subexcitation regime.



FIG. 5. Two views of the cumulative degradation spectral surface for subexcitation electrons generated, between energies of 0.5 and 12 eV, by the boundary term $Z(T_u, t)$ in argon at STP. Note that the condition in Eq. (14b) implies that this discontinuous maximum in $Z_{sh}(T,t)$ traces a curve in the *T*-*t* plane equivalent to T=g(t).

shown in two views in Figs. 5(a) and 5(b). [We recognize from Eq. (14b) that $Z_{sh} = 0$ for $t < \tau$.] Figure 5 shows that Z_{sh} experiences a sharp increase to a maximum at lower electron energies. At long times, the maximum prevails at the lowest energy. This behavior is attributable primarily to the presence of S(T) in the denominator of Eq. (14b).

We begin the calculation of Z_s in argon by first computing the corresponding steady-state yield Y_s . In the present case, Y is simply given by Eq. (12), which is plotted on a logarithmic scale in Fig. 6. The two quantities, $S(T_u)$ and $Z(T_u, \infty)=0.0043492$ cm/eV, used in evaluating Eq. (12) are the ones employed in generating Z_{sh} by Eq. (14b). The time limits and scale for the evaluation of Eq. (14b) were determined by first computing the slowing down time τ and its reciprocal g, according to Eqs. (13a) and (13b). The same two functions are also required for evaluation of Eqs. (14c) and (14d). The slowing down time τ for any gas at STP with a stopping cross section S, as plotted in Fig. 4, is displayed in Fig. 7. The energy interval chosen, 0.5 eV (about 20 times thermal energy) to 12 eV, is approximately the onset of electronic excitation in argon. As Fig. 7 shows, the slowing-down interval from 12 to 0.5 eV is approximately 350 ns. Moreover, the behavior of $\tau(T)$ is dominated by the dramatic variation of S over the same energy interval.

Once the numerical integrations are carried out in Eqs. (14c) and (14d) Z_s is obtained by superposition according



FIG. 6. $Y_s(T)$, as defined by Eq. (12), which is the steadystate solution of Eq. (10) for subexcitation electrons generated at a constant integrated rate of 1 eV⁻¹ by 1000-eV electrons in argon at STP.



FIG. 7. Equation (13), slowing-down time, from 12 to 0.5 eV for subexcitation electrons generated at a constant integrated rate of 1 eV⁻¹ by 1000-eV electrons in argon at STP, with the stopping power of Fig. 4.



FIG. 8. Two views of the cumulative degradation spectral surface, Eq. (14a), for subexcitation electrons generated at a constant integrated rate of 1 eV^{-1} by 1000-eV electrons. The degradation range is electron energies of 0.5-12 eV in argon at STP. The stopping cross section S(T) of the medium gas is the one displayed in Fig. 4. Note that the maximum in $S(T)Z_s(T,t)$ traces a curve in the *T*-t plane that is equivalent to $T \approx g(t)$.

to Eq. (14a) (although Z_{sh} makes a minor contribution to the overall Z_s). This surface is displayed from two perspectives in Fig. 8. The near view, Fig. 8(a), shows how the maximum in Z_s evolves from higher subexcitation energies. Both Figs. 8(a) and 8(b) show that the maximum experiences a sharp excursion to longer times at lower energies. This maximum mirrors the behavior of $\tau(T)$, as depicted in Fig. 7. As mentioned in the discussion of Eq. (14), a trace of the maximum in the quantity $S(T)Z_{sp}(T,t)$ follows g(t) in the T-t plane, a fact that is qualitatively illustrated by the behavior exhibited in Fig. 8.

III. CONCLUSION

We have outlined a method for deriving the timedependent degradation spectrum for subexcitation electrons from the degradation spectrum computed for highenergy electrons. We have also demonstrated the utility of this method in the case of an exactly calculable model, as well as in the real case of electrons slowing down in argon. The dominant characteristic of the cumulative degradation spectrum is the existence of a maximum in the SZ- τ plane at $\tau \approx t$ or in the Z_s -T plane at $T \approx g(t)$. Hence, the projection of the maximum onto the T-t plane traces a curve corresponding approximately to the energy dependence of the CSDA time displayed in Fig. 7. The present method permits the use of existing data to analyze the results of experiments that monitor the products of subexcitation electrons generated by ionizing radiation. This application was amply demonstrated by the use of experimentally determined argon inelastic and momentum transfer cross sections and a previously calculated Z surface to construct nR of Eq. (18). This source function, in turn, was used to characterize the cumulative subexcitation electron yield via Eq. (10). If Z_s is perturbed by a small amount of an additive that scavenges subexcitation electrons from the system, Eq. (10) can be modified by the addition of a homogeneous term, as in Ref. [20].

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- M. Inokuti, M. A. Dillon, and M. Kimura, Int. J. Quantum Chem. Symp. 21, 251 (1987).
- [2] M. Inokuti, M. Kimura, and M. A. Dillon, Phys. Rev. A 38, 1217 (1988).
- [3] M. Inokuti, M. Kimura, and M. A. Dillon, Radiat. Phys. Chem. 34, 477 (1989).
- [4] K. Kowari, Phys. Rev. A 41, 2500 (1990).

- [5] K. Kowari, M. Inokuti, and M. Kimura, Phys. Rev. A 42, 795 (1990).
- [6] M. Kimura, I. Krajcar, T. Teng, and M. Inokuti, Phys. Rev. A 46, 2532 (1992).
- [7] M. Kimura, M. A. Dillon, and M. Inokuti, Adv. Chem. Phys. 84, 193 (1993).
- [8] R. L. Platzman, Radiat. Res. 2, 1 (1955).

- [9] R. Cooper and M. C. Sauer, Jr., J. Phys. Chem. 93, 3 (1989).
- [10] R. Cooper and M. C. Sauer, Jr., J. Phys. Chem. 93, 1881 (1989).
- [11] R. Cooper, L. S. Denison, and M. C. Sauer, Jr., J. Phys. Chem. 86, 5093 (1982).
- [12] C. Naleway, M. Inokuti, M. C. Sauer, and R. Cooper, J. Phys. Chem. 90, 6154 (1986).
- [13] L. S. Denison, R. Cooper, and M. C. Sauer, Jr., J. Phys. Chem. 90, 683 (1986).
- [14] R. Cooper and M. C. Sauer, Jr., Radiat. Phys. Chem. 32, 34 (1989).
- [15] K. Kowari, L. Demeio, and B. Shizgal, J. Chem. Phys. 97,

2061 (1992).

- [16] K. Kowari and B. Shizgal, Appl. Radiat. Isot. 42, 985 (1991).
- [17] M. Inokuti, M. Kimura, and K. Kowari, Chem. Phys. Lett. 153, 504 (1988).
- [18] M. Kimura, M. Inokuti, and K. Kowari, Phys. Rev. A 40, 2316 (1989).
- [19] M. Inokuti, *Molecular Processes in Space* (Plenum, New York, 1990), p. 65.
- [20] M. A. Dillon, M. Inokuti, and M. Kimura, Radiat. Phys. Chem. 32, 43 (1988).
- [21] E. Eggarter, J. Chem. Phys. 62, 833 (1975).
- [22] M. Hayashi (private communication).