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Cross sections for L x-ray production and L-subshell ionization by MeV electrons

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A systematic study of L x-ray production and L-subshell ionization by MeV electron impact has been made. Cross sections were measured for 14 elements from Ba ($Z = 56$) to Bi ($Z = 83$) at electron bombarding energies of 1.04, 1.39, and 1.76 MeV. Within the error limits there is no dependence on the electron energy. At all energies, measured L x-ray production cross sections exhibit a gradual increase with atomic number. For the heavier elements, typical average values for the $L\alpha$, $L\beta$, and $L\gamma$ group are 70, 55, and 9 b, respectively. L-subshell ionization cross sections were derived from the $L\alpha$, $L\beta$, and $L\gamma$ x-ray production cross sections with the use of theoretical L-subshell fluorescence yields, Coster-Kronig yields, and radiative branching ratios. The total L-shell ionization cross section drops from 1200 b at Ba ($Z = 56$) to 380 b at Bi ($Z = 83$) with individual L-subshells contributing roughly in proportion to their electron number. The measured cross sections are in good agreement with the predictions of the binary-encounter collision model of Gryziński.

I. INTRODUCTION

Experiments on inner-shell ionization by electron impact have in the majority of cases been restricted to K-shell ionization at both nonrelativistic[1,2] and relativistic[3] energies. Very few data are available on ionization in the L shell. Relative L-shell ionization cross sections have been measured for electron energies up to several times the ionization energy for the gold L_2 and L_3 subshells,[4] the tungsten L_1, L_2, and L_3 subshells,[5] and the silver L_3 subshell.[6] Absolute inner-shell ionization cross sections have been determined only for the three L subshells of gold for electron energies up to about 40 keV.[7] In the extreme relativistic range of electron energies, absolute L x-ray production cross sections have been measured from 150 to 900 MeV for Tm, Ta, Au, and Bi.[8] In that work,[8] no attempt was made to derive the L-shell ionization cross sections because the Coster-Kronig transition yields and the three L-subshell fluorescence yields were not well known at that time. Up to the present, no systematic study of L-shell ionization by electron impact for a range of elements has been performed.

Theoretical treatments of inner-shell ionization by electrons are in the majority of cases nonrelativistic.[1] Furthermore, with one exception[9] the nonrelativistic calculations are for K-shell ionization only. This latter restriction also applies to most relativistic calculations.[1,10] The only theoretical treatment that is directly applicable to the present experiment is the classical binary collision model of Gryziński,[11] suitably modified to account for relativistic effects.

In this paper we will present the results of L x-ray production and L-subshell ionization measurements for 14 elements ranging from Ba ($Z=56$) to Bi ($Z=83$). Electron impact energies of 1.04, 1.39, and 1.76 MeV were used. A brief report of this work has been given previously.[12]

II. EXPERIMENTAL

A detailed description of the experimental procedure and setup is contained in an earlier paper on K-shell ionization by 2-MeV electron impact[3] which will hereafter be referred to as I. A brief review of the relevant information is given below.

A. Procedure

Relative x-ray production cross sections for the $L\alpha$, $L\beta$, and $L\gamma$ groups are determined from a measurement of the corresponding relative x-ray intensities after suitable corrections for absorption and relative detector efficiency. For this purpose thin targets containing a known atom ratio of the element to be measured and the reference element Cd were prepared. The relative intensity for a particular x-ray was obtained from its counting rate relative to the Cd $K\alpha$ x-ray and the atom ratio. Cd was chosen as the reference element because of its convenient chemical properties and because its $K\alpha$ x ray with an energy of 23 keV is not easily absorbed and does not interfere with the measurement of the L x rays.

The fluorescent x rays following electron impact were observed with a semiconductor Ge(Li) spectrometer and stored on a PDP 15/20 on-line computer. X-ray intensities were derived from the observed spectra in the following fashion. First, a background bremsstrahlung spectrum was obtained with a clean piece of the same thin lens tissue paper that was used as target substrate (see Sec. II D) in place at the target position. This background spectrum was matched to the observed spectrum in the flat region just above the Cd *K* lines and subtracted. The Cd $K\alpha$ and $K\beta$ x-ray intensities were then obtained by summing over appropriate channels. Second, the escape peaks of the Cd *K* lines, as obtained from a reference spectrum through a scaling procedure based on the Cd $K\alpha$ x-ray intensity, were subtracted channel by channel. Next, the background spectrum was matched once more above the *L* x-ray region and subtracted. This last step was necessary to account for the slight dependence of the slope of the bremsstrahlung background on the target material. Finally, the intensities of the *L* x-ray groups were determined by summing over appropriate channels.

B. Setup

The setup was essentially identical to the one described in I. Electrons accelerated with the 4-MeV Dynamitron accelerator at the State University of New York at Albany struck a target mounted with its normal at 45° with respect to the direction of the incident beam. Three different electron energies, 1.04, 1.39, and 1.76 MeV were used for all targets. The estimated energy error was ±0.05 MeV. X rays from the target were observed at 90° with respect to the beam direction. Spectra were typically accumulated for a total charge of 20 μC at average beam currents of 0.5 μA.

C. X-ray detection system

The same Ge(Li) detector, placed 41.6 cm from the target, and the same shielding arrangement as in I were used. However, the air path from the 0.051-mm-thick Mylar exit window to the detector front face was reduced to 3.5 cm by inserting a 2.54-cm diameter evacuated brass pipe between target chamber and detector. In addition, a 2.8-mm-diam. brass collimator was inserted in front of the detector window to improve detector resolution and line shape.

For the range of x-ray energies considered in the present experiment (up to 26 keV for the Cd $K\beta$), the total detection efficiency of the Ge(Li) detector is unity. To determine the photopeak efficiency, escape peak ratios as defined by Israel, Lier, and Storm[13] were measured for the $K\alpha$ lines of 11 elements ranging from $Z=34$ to 75 (Fig. 1). From these points, numerical values for the escape peak ratio were interpolated for each x-ray energy required.

Absorption of x rays between target and detector occurs in the Mylar exit window, the 3.5-cm-long air path, the 0.15-mm-thick Be window, and the 40.4-μg/cm^2 Au layer in front of the Ge(Li) detector. Absorption corrections were computed for

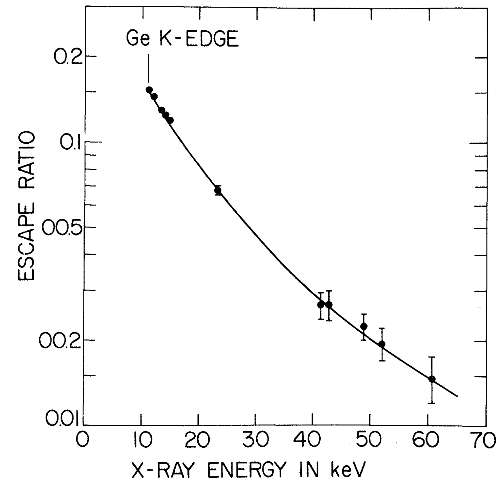

FIG. 1. Escape ratio $R=N_{esc}/(N_{ph}+N_{esc})$ as a function of the photon energy. N_{ph} and N_{esc} are, respectively, the count rates in the photo and both escape peaks. The smooth line represents a best fit through the data points.

each x-ray energy. For this purpose, the narrow-beam absorption coefficients of Storm and Israel[14] were interpolated. Their calculated values are in good agreement with experimental data[15] in our region of interest. The attenuation coefficients for Mylar and air were calculated from those of their components. The absorption correction is quite large for $Z\leq 60$. For instance, 51% of the Ba $L\alpha$ is absorbed, the major contributions being due about equally to the air path and the Mylar window. For $Z\geq 62$ the correction is always less than 30%.

D. Target and target corrections

The target preparation procedure was identical to the one described in I. Solutions with a known Z/Cd atom ratio were absorbed onto thin lens tissue paper (Fisher No. 11-996). The total target thickness obtained in this fashion is ~2.5 mg/cm^2 approximately half of which is due to the lens tissue backing. The validity of the target preparation procedure has been checked and reported on in detail in I.

For incident electrons in the MeV range, the energy loss in the target is less than 10 keV. Thus, the targets can be considered as thin and the x-ray production intensity is uniform throughout the thickness of the target. Excitation by bremsstrahlung produced in the target amounts to at most 1% of the *L*-shell ionization events as can be shown by a calculation similar[16] to the one in Appendix A of I. Excitation by characteristic x rays produced in the target can be estimated to less than a few tenths of one percent for the most unfavorable case, which is the excitation of the

$L\alpha$ by the $L\gamma$ x ray of the same element. Self-absorption of the low-energy x rays in the target, however, represents a significant correction and has to be considered. For this purpose an effective attenuation coefficient was measured for each x-ray line from each target by placing an identical target as absorber between target and detector. The fractional target transmission T, defined as the ratio of x rays emerging from the target to the number of x rays produced in it, was then calculated for the actual target geometry from Eq. (2) in I. Values for T range from 0.65 for the Ba $L\alpha$ to 0.98 for the Bi $L\beta$.

III. RESULTS AND DISCUSSION

A. X-ray production cross sections

The measured x-ray intensities were corrected for the fractional target transmission and for absorption in the path from the target to the detector. No corrections were made for the angular dependence of the emitted L x rays which, as is the case for K x rays,[8,17] was assumed to be isotropic. From the corrected relative intensities, x-ray production cross section were derived by dividing out the photopeak efficiency of the Ge(Li) detector and normalizing our relative measurements to the absolute $K\alpha$ x-ray production cross section for Cd of 28.3, 30.7, and 31.0 b at 1.04, 1.39, and 1.76 MeV, respectively. The latter values were obtained as in I from a subsidiary measurement with a Sn/Cd target by normalizing to the known absolute K-shell ionization cross section of Sn.[18] This normalization introduces a systematic error of ±10% on all our cross-section data.

The results for the $L\alpha$, $L\beta$, and $L\gamma$ x-ray production cross sections measured at the three electron impact energies are shown in Fig. 2. The numerical values obtained at 1.39-MeV electron impact energy are listed in Table I. Also shown in Fig. 2 are the total L x-ray production cross sections. These have been derived by summing the above three cross sections and adding a small correction of ~2% for the Ll x ray [obtained from $L\alpha$ using Eq. (2) below]. Over the range of elements studied in this experiment, there appears to be little variation with the electron impact energy. There is a gradual increase of the L x-ray production cross sections with Z. This behavior is opposite to the one observed for K x rays.[3,8] Also shown in Fig. 2 are representative errors, excluding the systematic error due to the normalization which could cause a parallel displacement of an entire set of data at one electron impact energy. These nonsystematic errors are estimated at 10% for $Z \leq 60$ and 5% for $Z \geq 62$. They are caused by background subtraction, uncertainties in the target transmission, and absorption corrections, as well as in the atom ratios of the targets. For the lower Z values, the poor resolution of the x-ray peaks from each other is also a contributing factor.

The connection of the x-ray production with the ionization cross sections is rather more involved for the L shell than for the K shell. In addition to electron impact ionization, vacancies in the L subshells can be produced by Auger and radiative transitions to the K shell, and by Coster-Kronig transitions between the L subshells. In analogy with Ref. 19 we derive vacancy production cross sections σ_i^v from the corresponding ionization cross section σ_i. The subscript i is used to distinguish the three L subshells:

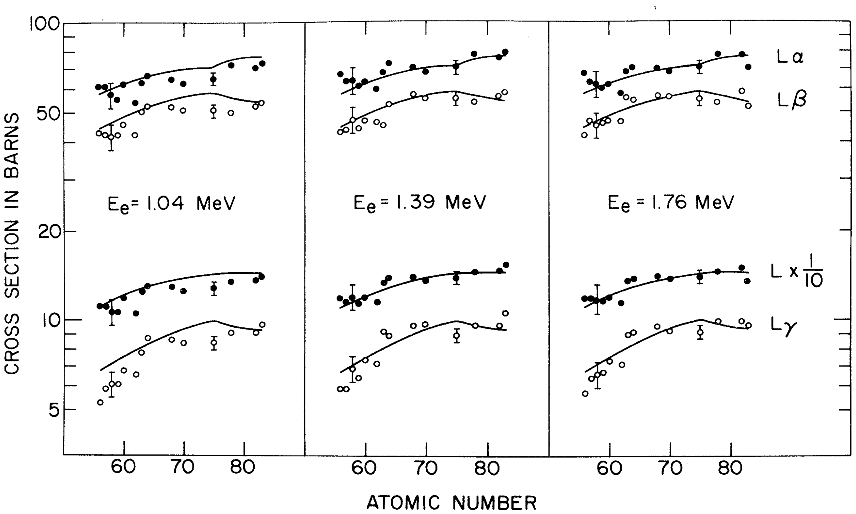

FIG. 2. $L\alpha$, $L\beta$, $L\gamma$, and total L x-ray production cross sections as a function of atomic number for 1.04-, 1.39-, and 1.76-MeV incident electron energy. Representative errors not including a systematic error of 10% resulting from the absolute normalization are indicated by bars for the regions $Z \leq 60$ and $Z \geq 62$. The solid curves are the result of a calculation using Gryziński's (Ref. 11) ionization cross sections. The kinks at $Z = 75$ are caused by discontinuities in the Coster-Kronig yields.

TABLE I. Measured $L\alpha$, $L\beta$, and $L\gamma$ x-ray production cross sections in b at 1.39-MeV electron impact energy. See text for a discussion of the normalization and the error.

Z	σ_α	σ_β	σ_γ
56	67.1	42.8	5.9
57	63.3	43.7	5.9
58	63.6	47.0	6.9
59	61.0	44.3	6.4
60	62.9	46.8	7.4
62	59.3	46.3	7.2
63	67.7	54.8	9.2
64	72.6	53.1	8.9
68	70.4	57.3	9.6
70	67.9	55.5	9.7
75	70.2	55.3	8.9
78	77.5	53.8	9.6
82	75.8	56.6	9.6
83	79.1	58.3	10.6

$$\begin{aligned} \sigma_1^v &= \sigma_1 + n_{K1}\sigma_K \ , \\ \sigma_2^v &= \sigma_2 + n_{K2}\sigma_K + f_{12}\sigma_1^v \ , \\ \sigma_3^v &= \sigma_3 + n_{K3}\sigma_K + f_{13}\sigma_1^v + f_{23}\sigma_2^v \ . \end{aligned} \tag{1}$$

Here, n_{Ki} is the average number of vacancies produced in the L_i subshell by Auger and radiative transitions to one K-shell vacancy, and f_{ij} is the Coster-Kronig yield for shifting a vacancy from the ith to the jth L subshell. Numerical values for the n_{Ki} have been obtained from a fit to the available experimental information.[20] To obtain the f_{ij} for the elements measured, we have interpolated the theoretical values given in Ref. 21. σ_K is the experimental K-shell ionization cross section which has been remeasured as described in I for the electron bombarding energies relevant to this experiment and found to be in substantial agreement with the results presented in I.

The probability that a vacancy in the L_i subshell is filled via a radiative transition is given by the fluorescence yield ω_i. However, unlike the case of K x rays, the $L\beta$ and $L\gamma$ x-ray groups are composed of several transitions, which cannot be resolved with a Ge(Li) detector. To complicate matters further, the composite transitions terminate on more than one of the L subshells. Figure 3 indicates in a schematic fashion the most intense constituent lines as identified by Bearden[22] and Scofield.[23] In order to calculate the x-ray production cross sections, we define radiative branching fractions F_{ij}. As for instance, $F_{3\alpha}$ is the ratio of the sum of the transition rates of the L_3-M_4 and L_3-M_5 transitions, which form the $L\alpha$ x-ray group, to the total transition rate to the L_3 subshell. Other possible branching fractions are defined in an analogous fashion. These branching fractions have been calculated from the theoretical individual transition rates to the L subshells.[23]

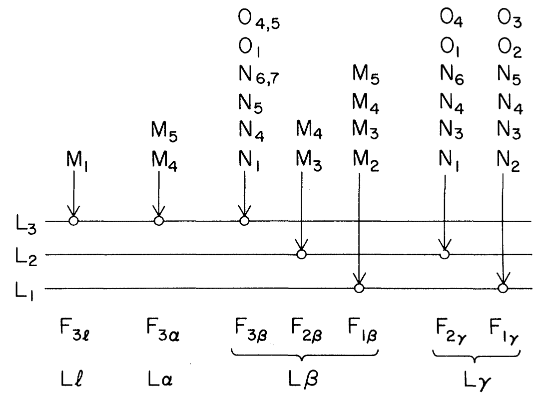

FIG. 3. Schematic representation of x-ray transitions contributing to radiative branching fractions F_{ij} and observed unresolved L x-ray groups. For details see text.

With the above definitions we can relate the L x-ray production cross sections to the L-subshell vacancy production cross sections:

$$\begin{aligned} \sigma_l &= \sigma_3^v \omega_3 F_{3l} \ , \\ \sigma_\alpha &= \sigma_3^v \omega_3 F_{3\alpha} \ , \\ \sigma_\beta &= \sigma_3^v \omega_3 F_{3\beta} + \sigma_2^v \omega_2 F_{2\beta} + \sigma_1^v \omega_1 F_{1\beta} \ , \\ \sigma_\gamma &= \sigma_2^v \omega_2 F_{2\gamma} + \sigma_1^v \omega_1 F_{1\gamma} \ . \end{aligned} \tag{2}$$

Using Eqs. (1) and (2), the x-ray production cross sections can be calculated from the L-subshell ionization cross sections. The theoretical values of Ref. 24 have been interpolated to obtain the L-subshell fluorescence yields for the elements measured. The solid lines in Fig. 2 have been obtained using Gryziński's[11] theory to predict the ionization cross sections. In the latter calculation, experimental values for the ionization energies[25] have been used. The agreement with the experimental data is rather remarkable. With the exception of the data at 1.04-MeV electron impact energy, which seem systematically low and may reflect a normalization error, theory and experiment generally overlap within the (nonsystematic) error limits. This not only indicates that the Gryziński[11] model accurately predicts the ionization cross section. It also implies that the largely theoretical estimates of those other quantities, that substantially enter into the calculation of the L x-ray production cross sections, are essentially correct. Of course, the presence of mutually compensating errors of 10 to 20% magnitude in the various theoretical estimates cannot be excluded on the basis of our experimental data. While there is general agreement between theoretical and experimental values for the fluorescence and Coster-Kronig yields, experimental errors are still large, especially for the Coster-Kronig yields.[19] No experimental values are available for the radiative branching fractions used in the present analysis. However, selected transition probability ratios have been measured[26] for a number of elements with $Z \geq 57$. While systematic discrepancies with theory[23] have been uncovered, they are less than 22% for the most unfavorable case.[26]

The present experiment does not distinguish between single- and multiple-ionization events as long as the lowest vacancy is created in the L shell. For electron impact, the shakeoff model usually provides reasonably accurate estimates for the relative probability of multiple ionization[27] even in cases[28] where multiple Coulomb ionization[11] is also present. In as much as shakeoff follows the primary ionization process, the theoretical single-ionization cross sections include multiple ionization and thus can be directly compared to the experiment. Changes in the average Coster-Kronig and fluorescence yields due to the presence of multiple vacancies are expected to be small, since the shakeoff probability is spread over many subshells. For instance, theoretical estimates[29] for Xe indicate probabilities of $\lesssim 2.5\%$ for all subshells that are involved in the more intense L x-ray transitions.

B. L-subshell ionization cross sections

The extraction of accurate values for the L-subshell ionization cross sections from the experimental x-ray production cross sections is made cumbersome by the uncertainties on some of the factors occurring in Eqs. (1) and (2). Furthermore, the determinants that appear in the solution of Eq. (2) are nearly singular. To circumvent these problems, a χ^2 procedure was followed. χ^2 was obtained from a comparison of experimental and calculated x-ray production cross sections. For this purpose, χ^2 was minimized for each element by varying the ionization cross sections about the Gryziński[11] values with the ratio σ_1/σ_2 held at a constant ratio a, independent of Z. This condition is in qualitative agreement with the behavior of the theoretical ionization cross sections and appears reasonable since it does not seem likely that this cross section ratio will undergo abrupt changes from one element to the next. All other quantities in Eqs. (1) and (2) are taken as defined previously. Errors in these quantities were neglected in this analysis since their magnitude is not well known.

The sum of the minimized χ^2 values for all 14 elements, as a function of a, exhibits a rather shallow minimum with fairly steep walls restricting a to the range $0.70 \lesssim a \lesssim 1.20$. The Gryziński model yields $a \approx 0.94$. The ionization cross sections corresponding to the χ^2 minimum for each individual element are shown in Fig. 4. The points and the error bars for σ_1 and σ_2 correspond to, respectively, the Gryziński and the extreme values for a as given above. These errors are, of course, correlated. In comparison, errors resulting from uncertainties in the measured x-ray production cross sections are small for these two subshells, and they are therefore not shown. The L_3-subshell ionization cross section σ_3 as well as the total L-shell ionization cross section, on the other hand, exhibit only a minor dependence on the choice of a. Their typical errors for the regions $Z \leq 60$ and $Z \geq 62$ have been arrived at from the errors in the x-ray production cross sections in Fig. 3. The systematic error due to the normalization is not shown. Again, because of the linear form of Eqs. (1) and (2), its effect could be only a parallel displacement of all data points. The solid curves are calculated from Gryziński's theory.[11] There is generally agreement with the experimental points within the error limits.

It is somewhat surprising that the Gryziński theory,[11] which is essentially a classical binary collision model modified for relativistic effects, should lead to such good agreement with the experiment. The Kolbenstvedt theory,[10] which incorporates quantum-mechanical aspects, has unfortunately not been extended to the case of L-shell ionization. In I it had been shown that the latter theory is in rather good agreement with the general trend of the K-shell ionization data at 2-MeV electron impact energy, but that predicted cross sections are systematically too large in the high-Z region. An application of Gryziński's model to this experiment shows close agreement between the two competing theories in the low-Z region. In the high-Z region, however, the Gryziński model gives better results in that it does not show the systematic deviation from the trend of the experimental data.[30] Kolbenstvedt's theory, on the other hand, is superior in the extreme relativistic range of electron energies from 150 to 900 MeV.[8] At these energies, the experimental cross sections are by about a factor of 2.5 to 3.0 higher than at 2 MeV. While Gryziński's model does exhibit such a relativistic rise in the cross section, it can only account for half of the observed increase. In contrast, Kolbenstvedt's theory can account for it fully.[8] It should be noted, however, that this close agreement with the experiments at ultrarelativistic electron energies did require modifications[8] on the

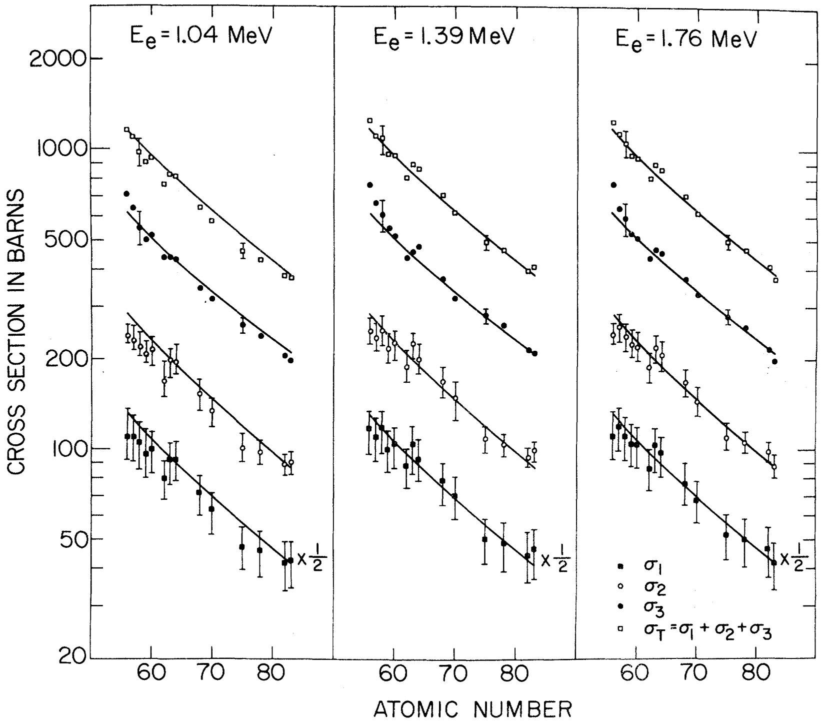

FIG. 4. L-subshell ionization cross sections as a function of atomic number for 1.04-, 1.39-, and 1.76-MeV incident electron energy. The solid curves are calculated from Gryziński's (Ref. 11) binary collision theory. For a discussion of the error bars see text.

original calculation.[10] Without these modifications, the Kolbenstvedt model[10] also underestimates the cross sections, although only for the case of the low-Z elements. It would be very desirable to have a better understanding of the limitations underlying the two competing models.

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