

## Cross Section for $M$ -Shell Ionization in Heavy Atoms by Collision of Simple Heavy Charged Particles\*

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The cross sections for  $M$ -shell electron ionization by direct Coulomb excitation of heavy target atoms by incident heavy charged particles are evaluated. Incident particles are described in the plane-wave Born approximation, and screened hydrogenic wave functions are used for the atomic electrons. Numerical results are given for Au, U, and No, and the theory is compared with experimental data for Ho as an illustration. Explicit expressions for the absolute values of the  $M$ -subshell form factors of bound-free transitions are presented as functions of momentum and energy transfers. The results are discussed.

### I. INTRODUCTION

The inner-shell ionization of atoms by incident heavy charged particles such as protons or  $\alpha$  particles has been a process of interest for both atomic and nuclear physicists in connection with the production of characteristic x rays following the creation of vacancies in inner shells.<sup>1</sup> Calculations of the ionization cross section by direct Coulomb excitation have usually been made based on the plane-wave Born approximation (PWBA) both for  $K$  and  $L$  shells.<sup>2,3</sup> In this approximation scheme, incident and inelastically scattered particles are described by plane waves using first-order perturbation theory. Incident particles are assumed to be bare charged particles and the electronic structures are neglected. Furthermore, screened hydrogenic wave functions have commonly been used for the atomic electrons. These assumptions of the PWBA scheme restrict the applicability of calculations in terms of incident energies, charge of the projectiles, and target atoms. For very low incident energies such that  $zZe^2/\hbar v \gg 1$ , the PWBA predictions on the  $K$ -shell ionization cross sections disagree with the experimental data by an order of magnitude,<sup>4</sup> where  $ze$  and  $v$  are the charge and velocity of the projectile and  $Z$  is the charge number of target atomic nucleus. Moreover, for incident energies less than 0.1 MeV/amu, the interpretation of inner-shell vacancies in terms of Coulomb excitation may not be appropriate.<sup>5</sup>

However, the PWBA predictions based on the use of screened hydrogenic wave functions of atomic electrons provide good agreement with the experimental  $K$ -shell ionization cross sections and agree reasonably well with the experimental  $L$ -shell ionization cross sections for  $Z \geq 30$  for incident energies satisfying  $zZe^2/\hbar v \lesssim 1$ . For  $M$  electrons, the above condition for the energies of incident particles could be relaxed, since the velocities of

$M$  electrons are slower than those of  $K$  and  $L$  electrons, while the use of screened hydrogenic wave functions restricts the choice of target atoms to heavier elements. For heavy target atoms ( $Z \geq 75$ ) such as gold, lead, or the transuranic elements, the  $M$  shell may also be considered to be an inner shell, and screening effects due to outer electrons are less important compared to other lighter elements. Recently,  $M$  x-ray-yield cross sections for heavy elements have been measured.<sup>6,7</sup> In this paper,  $M$ -shell ionization cross sections have been evaluated based on the PWBA scheme, following the previous work on  $K$ - and  $L$ -shell ionization-cross-section calculations.<sup>2,3</sup> These calculations are intended to provide reasonable estimates of the absolute values and of the expected energy dependence of the cross section for  $M$  x-ray production due to collisions of protons or  $\alpha$  particles with heavy target atoms.

The method is outlined in Sec. II presenting explicit expressions for  $M$ -shell form factors for the transition from each  $M$  subshell to the continuum final states. Expressions of the  $M$ -shell form factors given earlier by Khandelwal *et al.*<sup>8,9</sup> are found to be in error, although their numerical calculation of the stopping power of  $M$  electrons is in reasonable agreement with those obtained from the new form factors reported in this paper.

Results of the present calculation on the  $M$ -shell ionization cross section for some specific heavy elements (Au, U, and No) are presented for comparison with the experimental data.  $M$ -shell ionization cross sections for medium heavy elements such as Ho are also presented and compared with the experimental data for purpose of illustration. The results are discussed in Sec. III.

### II. $M$ -SHELL IONIZATION CROSS SECTION

The ionization cross section of electrons ejected from the  $M_i$  subshell by incident heavy charged particle is given by<sup>2</sup>

$$\sigma_{M_i} = \frac{8\pi Z^2 a_0^2}{Z_{M_i}^4 \eta_{M_i}} \int_{W_{\min}}^{\infty} dW \int_{Q_{\min}}^{\infty} \frac{dQ}{Q^2} |F_{W, M_i}(Q)|^2, \quad (1)$$

$$\eta_{M_i} = mE/MZ_{M_i}^2 R_{\infty}, \quad W = \Delta E/Z_{M_i}^2 R_{\infty},$$

$$Q = q^2 a_0^2 / Z_{M_i}^2 \quad (i = 1, 2, 3, 4, 5)$$

in the nonrelativistic plane-wave Born approximation (PWBA), where  $M_{1,2,3,4,5}$  refer to  $3s_{1/2}$ ,  $3p_{1/2}$ ,  $3p_{3/2}$ ,  $3d_{3/2}$ ,  $3d_{5/2}$  atomic subshells, respectively.  $M$  and  $E$  are the mass and energy of the incident particle,  $\Delta E$  and  $\hbar\vec{q}$  are the energy and momentum transfers of the incident particle to the  $M_i$  electrons, and  $Z_{M_i}$  is the effective-nuclear-charge number seen by the  $M_i$  electrons.<sup>10</sup>

The form factor  $F_{W, M_i}(Q)$  for the transition between electronic states  $\psi_{M_i}(\vec{r})$  and  $\psi_W(\vec{r})$ , for the electron initially bound in the  $M_i$  subshell and finally ejected with energy transfer  $\Delta E$ , is defined to be

$$F_{W, M_i}(Q) = \int e^{i\vec{q} \cdot \vec{r}} \psi_{M_i}(\vec{r}) \psi_W^*(\vec{r}) d\vec{r}. \quad (2)$$

For the electronic wave functions  $\psi_{M_i}(\vec{r})$  and  $\psi_W(\vec{r})$ , we have used nonrelativistic screened hydrogenic wave functions.  $W_{\min}$  is the observed ionization potential  $I_{M_i}(Z)$  of the  $M_i$  subshell.<sup>11</sup> The screening effects due to outer electrons have been taken into account in the same way as in the earlier calculations of the *K*- and *L*-shell ionization cross sections.<sup>2,3</sup>

A calculation employing relativistic wave functions for atomic electrons would increase the *M*-shell ionization cross sections but would not affect the cross sections as much as in the case of *K*- and *L*-shell ionization cross sections,<sup>12,13</sup> for the velocities of *M* electrons, approximately given by  $Z_{M_i}e^2/3\hbar$ , are much less than  $c$ , the velocity of light. One might still take into account relativistic effects, while using the nonrelativistic screened hydrogenic wave functions, by assuming a corrected value for  $W_{\min} = I'_{M_i}(Z)$ , the effective ionization potential of an  $M_i$  subshell, as described in Merzbacher and Lewis.<sup>2</sup>  $I'_{M_i}(Z)$  is given as

$$I'_{M_i}(Z) = I_{M_i}(Z) (1 - a_{M_i}),$$

with

$$a_{M_i} = (I_R - I_{NR})/I_{M_i}(Z),$$

where  $I_R$  and  $I_{NR}$  are the relativistic and nonrelativistic ideal ionization potentials in the absence of outer screening in units of  $Z_{M_i}^2 R_{\infty}$ . Thus, from the Dirac theory of the hydrogenic atom,

$$I_R = \frac{2 \{1 - [1 + (Z_{M_i} \alpha)^2 / (n + \gamma_k)^2]^{-1/2}\}}{(Z_{M_i} \alpha)^2},$$

$$I_{NR} = \frac{1}{3}, \quad \gamma_k = [\kappa^2 - (Z_{M_i} \alpha)^2]^{1/2}, \quad \alpha = e^2/\hbar c.$$

$n + |\kappa| = 3$  for the *M* shell, and the angular momentum of electrons  $j = |\kappa| - \frac{1}{2}$ . The  $M_{3,4,5}$  subshells give the dominant contributions to the *M*-shell ionization cross sections, as discussed later. For example, applying the above relativistic corrections to the subshells of uranium ( $Z = 92$ ), we obtain  $a_{M_3} = 0.070$ ,  $a_{M_4} = 0.047$ , and  $a_{M_5} = 0.016$ , using the effective charge number  $Z_{M_i}$  given in Sec. III.

Heretofore, it has been assumed that *M*-shell ionization takes place in an atom which has all of its electrons filled. Observations of complex satellite structure suggest that this condition is frequently not satisfied and that one or more outer electrons are usually removed at the same time. In the presence of a hole in an outer shell, e.g., in  $N_j$  subshell,  $W_{\min} = I_{M_i(N_j)}(Z)$  and

$$I_{M_i(N_j)}(Z) = I_{M_i}(Z) + I_{N_j(M_i)}(Z) - I_{N_j}(Z) \\ \simeq I_{M_i}(Z) + I_{N_j}(Z+1) - I_{N_j}(Z) > I_{M_i}(Z).$$

Here,  $I_{A(B)}(Z)$  denotes the observed ionization potential of a *A*-subshell electron with a hole in *B* subshell, and an approximation  $I_{N_j(M_i)}(Z) \simeq I_{N_j}(Z+1)$  has been made.<sup>14</sup> For example,  $I_{M_5(N_1)}(Z) \simeq 1.017 I_{M_5}(Z)$  for uranium. Therefore, it will slightly reduce the ionization cross section. In this paper, we are mainly concerned with the neutral target atoms.

The algebraic manipulations required to obtain the  $M_i$ -subshell form factors employing the well-known scheme of Bethe<sup>15</sup> and Walske<sup>16</sup> are manageable only through the use of a computer. We present the results in the following form:

$$|F_{W, M'}(Q)|^2 = \frac{2^7 Q \exp\{- (2/k) \arctan[\frac{2}{3} k / (Q - k^2 + \frac{1}{9})]\}}{3^3 (1 - e^{-2\pi/k}) [(Q - k^2 + \frac{1}{9})^2 + \frac{4}{9} k^2]^2} \sum_{i, j=0}^{i_{\max}, j_{\max}} C_{ij}^{(M')} k^{2i} Q^j \quad (M' = 3s, 3p, 3d) \quad (3)$$

for the transition from initial  $3s$ ,  $3p$ ,  $3d$  states to the final continuum states characterized by  $W = \frac{1}{3} + k^2$ .

The  $M_i$ -subshell form factors  $|F_{W, M_i}(Q)|^2$  were obtained from the relation

$$|F_{W, 3i, j}(Q)|^2 = \frac{2j+1}{2(2l+1)} |F_{W, 3i}(Q)|^2$$

$$(l = 0, 1, 2 \text{ and } j = l \pm \frac{1}{2}),$$

which is valid in a nonrelativistic treatment that neglects the spin dependence of the atomic states.

The coefficients  $C_{ij}^{(M')}$  are presented in tabular form in the Appendix, which also includes the expression for

$$|F_{W, M}(Q)|^2 = \sum_{i=1}^5 |F_{W, M_i}(Q)|^2 = \sum_{M' = 3s, 3p, 3d} |F_{W, M'}(Q)|^2.$$

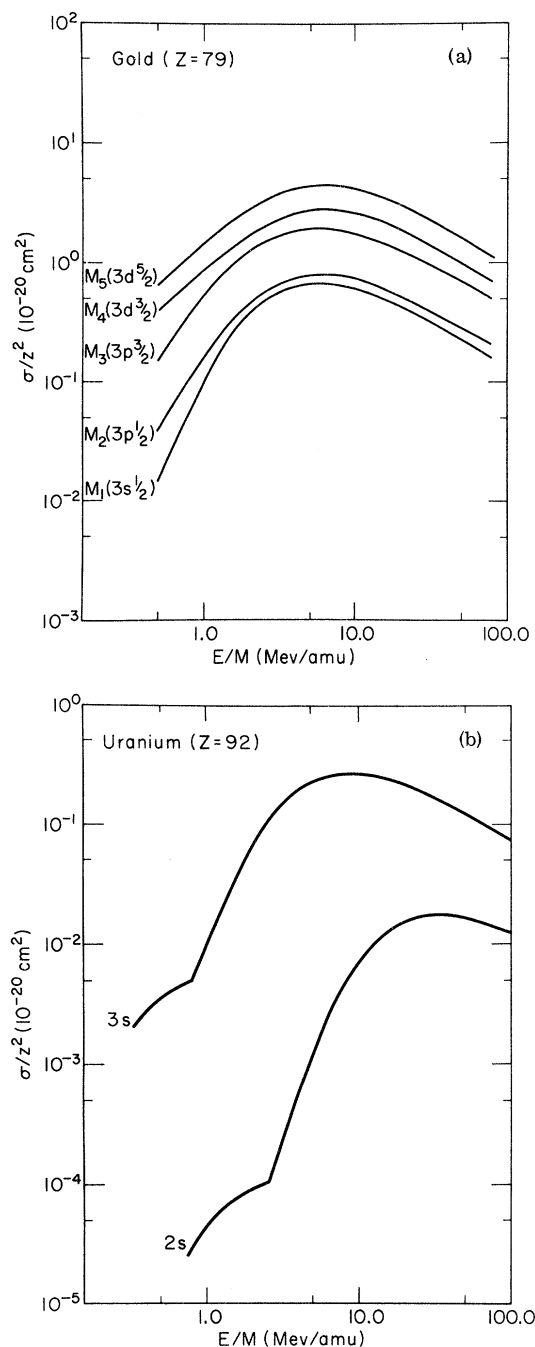


FIG. 1. (a)  $M_i$ -subshell ionization cross section  $\sigma_{M_i}/z^2$  for gold as a function of incident energy  $E/M$ . (b) Comparison between  $M_1(3s_{1/2})$ - and  $L_1(2s_{1/2})$ -subshell ionization cross sections,  $\sigma_{M_1}/z^2$  and  $\sigma_{L_1}/z^2$ , for uranium as a function of incident energy  $E/M$ .

The above expressions have been checked by substituting  $k = i/n$ , obtaining the expressions for the generalized hydrogenic oscillator strengths for the discrete transitions  $3s$ ,  $3p$ ,  $3d \rightarrow n$ .

Finally, the total  $M$ -shell ionization cross section

is given by  $\sigma_M = \sum_{i=1}^5 \sigma_{M_i}$ . The double integrations in Eq. (1) were performed numerically with the appropriate change of variables. Errors in the numerical computation of the present work are, in general, less than 1%.

### III. RESULTS AND DISCUSSION

Ionization cross sections for  $M$  electrons by incident heavy charged particles were evaluated for incident energies  $E/M = 0.5 - 100.0$  Mev/amu and for several species of heavy target atoms. Here,  $M$  denotes the mass of the incident particle.

For the effective screened charge number  $Z_{M_i}$  of the  $M_i$  electrons, according to Slater's rule,<sup>17</sup> we have used  $Z_{M_1} = Z_{M_2} = Z_{M_3} = Z - 11.25$  and  $Z_{M_4} = Z_{M_5} = Z - 21.15$  for definiteness.

In Fig. 1(a), the contributions of all  $M_i$  subshells to the total  $M$ -shell ionization cross section are shown for gold. Previously, Hansteen and Mosebakk<sup>18</sup> have evaluated the  $3s$ - and  $3p$ -subshell ionization cross sections for this element using a semiclassical impact-parameter approximation. Their calculations of the  $3s$  and  $3p$  contributions to the cross section gave results which are qualitatively similar but slightly larger in magnitude than the present calculations. It should be noted, however, that the dominant contribution to the total  $M$ -shell ionization cross section arises from the  $3d$  subshells as shown in Fig. 1(a). This is consistent with the fact that the  $3d$  subshell is the outermost shell among the  $M$  subshells.

A comparison between  $M_1(3s_{1/2})$ - and  $L_1(2s_{1/2})$ -subshell ionization cross sections is shown in Fig. 1(b). Sharp kinks in the lower-energy region are found in both subshell ionization cross sections. These can be attributed to the nodes in the momentum wave functions for  $3s$  and  $2s$  electrons.<sup>19</sup>

For purposes of illustration, in Fig. 2 the present calculation is compared with experimental data, taken from the work of Khan *et al.*,<sup>20</sup> on the ionization of the  $M$  shell of holmium ( $Z = 67$ ), a medium heavy element. Accurate experimental data on the average  $M$ -shell fluorescence yield  $\bar{\omega}_M$  do not exist for this element, and an order-of-magnitude estimate had to be used to determine the  $M$ -shell ionization cross section. Therefore, even though  $M$  x-ray yields were measured quite accurately, the  $M$ -shell ionization cross sections shown in Fig. 2 have large errors due to the indeterminacy of the  $M$ -shell fluorescence yield  $\bar{\omega}_M$ . Yet, the energy dependence of the present calculation agrees well with the experimental data, but the discrepancies in magnitude are appreciable. However, the present calculation is not expected to be reliable for medium heavy elements.  $M_4$ - and  $M_5$ -subshell ionization cross sections are also shown in Fig. 2. A different calculation on the  $M_4$ - and  $M_5$ -subshell ioniza-

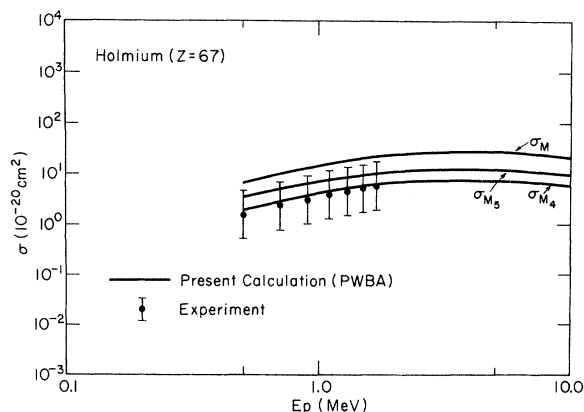


FIG. 2. *M*-shell,  $M_4(3d_{3/2})$ - and  $M_5(3d_{5/2})$ -subshell ionization cross sections,  $\sigma_M$ ,  $\sigma_{M_4}$ , and  $\sigma_{M_5}$ , for holmium compared with the experimental data as a function of incident proton energy  $E_p$ . Experimental data were taken from Ref. 20 and errors shown here are mainly due to indeterminacy of fluorescence yield.

tion cross section for this element has been published by Garcia,<sup>21</sup> who obtained the  $M_4$ - and  $M_5$ -subshell ionization cross sections using the classical binary-encounter approximation. His values are slightly larger than the present  $M_4$ - and  $M_5$ -subshell ionization cross sections.

Sample results of calculations on the *M*-shell ionization cross sections for heavy elements are presented in Table I for Au ( $Z=79$ ), U ( $Z=92$ ), and No ( $Z=102$ ) for comparison with experimental data, which also included the medium heavy element Ho for convenience.

In Figs. 3(a) and 3(b), two of these (Au and U)

TABLE I. *M*-shell ionization cross section  $\sigma_M/z^2$  as a function of incident energy  $E/M$ .

$E/M$ (MeV/amu)	$\sigma_M/z^2$ ( $10^{-20}$ cm $^2$ )			
	Ho ( $Z=67$ )	Au ( $Z=79$ )	U ( $Z=92$ )	No ( $Z=102$ )
0.5	6.705	1.296	0.228	0.069
1.0	14.878	3.310	0.676	0.244
1.5	20.651	5.267	1.176	0.439
2.0	24.221	6.869	1.684	0.649
3.0	27.383	8.987	2.569	1.068
4.0	27.896	10.081	3.214	1.432
6.0	26.254	10.706	3.932	1.935
8.0	23.843	10.458	4.195	2.205
10.0	21.575	9.930	4.231	2.331
12.0	19.612	9.337	4.155	2.371
14.0	17.944	8.757	4.026	2.361
16.0	16.526	8.219	3.876	2.323
20.0	14.268	7.287	3.563	2.205
25.0	12.197	6.363	3.203	2.037
30.0	10.670	5.643	2.895	1.876
35.0	9.501	5.071	2.648	1.731
40.0	8.577	4.606	2.428	1.603
50.0	7.208	3.900	2.081	1.394
60.0	6.238	3.391	1.821	1.231
70.0	5.513	3.007	1.620	1.102
80.0	4.949	2.705	1.461	0.997
100.0	4.124	2.262	1.225	0.840

are compared with the *L*-shell ionization cross sections. The maxima of the *L*- and *M*-shell ionization cross sections as a function of incident energy  $E/M$  occur around 40 and 10 Mev/amu, respectively, for uranium, while they are 25 and 6 Mev/amu for gold. These results may be understood in terms of the binding energies of *L* and *M*

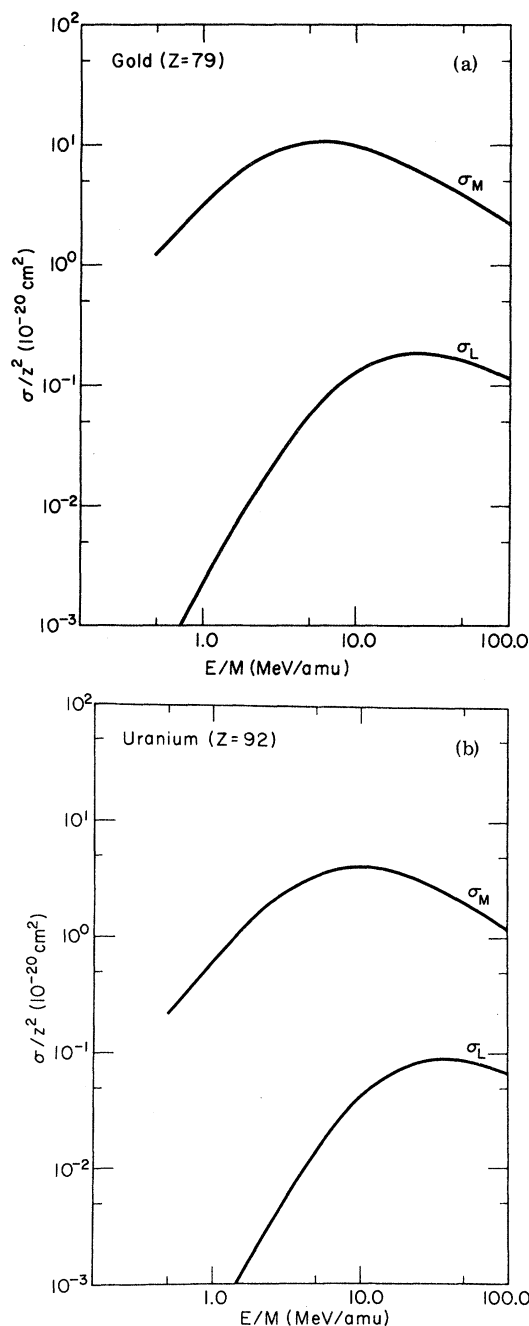


FIG. 3. Comparison between *M*- and *L*-shell ionization cross sections,  $\sigma_M/z^2$  and  $\sigma_L/z^2$ , for (a) gold and (b) uranium as a function of incident energy  $E/M$ .



TABLE IV. Coefficients  $C_{ij}^{(3d)}$ ,  $i_{\max} = j_{\max} = 7$ .

$j \backslash i$	0	1	2	3	4	5	6	7
0	$\frac{176}{3^{19}}$	$\frac{8816}{3^{19}}$	$\frac{6800}{3^{16}}$	$\frac{2800}{3^{13}}$	$\frac{17\ 680}{3^{13}}$	$\frac{2288}{3^{10}}$	$\frac{16}{3^5}$	$\frac{80}{3^7}$
1	$\frac{37\ 504}{5 \times 3^{18}}$	$\frac{473\ 504}{5 \times 3^{17}}$	$\frac{263\ 648}{5 \times 3^{14}}$	$\frac{660\ 544}{5 \times 3^{13}}$	$\frac{283\ 328}{5 \times 3^{11}}$	$\frac{18\ 784}{5 \times 3^8}$	$\frac{3616}{5 \times 3^7}$	
2	$\frac{656}{7 \times 3^{11}}$	$\frac{690\ 352}{5 \times 3^{15}}$	$\frac{318\ 688}{5 \times 3^{13}}$	$\frac{-218\ 912}{5 \times 7 \times 3^{10}}$	$\frac{-70\ 928}{5 \times 3^9}$	$\frac{-7184}{5 \times 3^7}$		
3	$\frac{3968}{5 \times 3^{12}}$	$\frac{-124\ 096}{3^{13}}$	$\frac{-579\ 392}{5 \times 3^{11}}$	$\frac{-33\ 344}{3^9}$	$\frac{-7744}{5 \times 3^7}$			
4	$\frac{-87\ 952}{5 \times 3^{12}}$	$\frac{222\ 224}{5 \times 3^{11}}$	$\frac{297\ 776}{5 \times 3^9}$	$\frac{16\ 496}{5 \times 3^7}$				
5	$\frac{5632}{5 \times 3^8}$	$\frac{-26\ 464}{3^9}$	$\frac{6176}{5 \times 3^7}$					
6	$\frac{208}{3^7}$	$\frac{-16}{3^2}$						
7	$\frac{512}{3^6}$							

measurements of *M*-shell x ray by Shafroth *et al.*,<sup>6,7</sup> although the absolute cross section has not been determined by these experiments. It will be interesting to compare the experimental data for heavy elements such as gold, lead, or transuranic elements with the present calculations.

It thus appears that the PWBA calculation of the *M*-shell ionization cross section, using simple screened hydrogenic wave functions, can give a reasonably good account of the production of an *M*-shell vacancy in heavy elements. However, more precise experimental measurements will call for more refined calculations employing better atomic wave functions.

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#### APPENDIX

The coefficients  $C_{ij}^{(M')}$  ( $M' = 3s, 3p, 3d$ ) in the expression of *M*-subshell form factor, Eq. (3), are

presented in Tables II–IV.

$|F_{W, M}(Q)|^2$  obtained by summing over all subshells  $M_i$  has a simple form:

$$|F_{W, M}(Q)|^2 = \frac{2^7 Q \exp\left\{-\frac{2}{k} \arctan\left[\frac{2}{3} k / \left(Q - k^2 + \frac{1}{3}\right)\right]\right\}}{3^3 \left[\left(Q - k^2 + \frac{1}{3}\right)^2 + \frac{4}{9} k^2\right]^{15} (1 - e^{-2\pi/k})}$$

$$\times [Q^5 + f_1(k)Q^4 + f_2(k)Q^3 + f_3(k)Q^2 + f_4(k)Q + f_5(k)],$$

with

$$f_1(k) = -\frac{43}{3^3} - \frac{11}{3} k^2,$$

$$f_2(k) = \frac{518}{3^5} + \frac{412}{3^4} k^2 + \frac{14}{3} k^4,$$

$$f_3(k) = -\frac{442}{3^6} - \frac{310}{3^4} k^2 - \frac{122}{3^3} k^4 - 2k^6,$$

$$f_4(k) = \frac{1943}{3^9} + \frac{1460}{3^7} k^2 + \frac{290}{3^5} k^4 + \frac{4}{3^3} k^6 - \frac{1}{3} k^8,$$

$$f_5(k) = \frac{377}{3^{11}} + \frac{2431}{3^{10}} k^2 + \frac{1790}{3^8} k^4 + \frac{62}{3^4} k^6 + \frac{71}{3^4} k^8 + \frac{1}{3} k^{10}.$$

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<sup>1</sup>For recent experimental data on the inner-shell ionization cross sections with improved resolutions, cf. *Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, Georgia* (Technical Information Division of the U.S.A.E.C., Oak Ridge, Tenn., 1973).

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