PITT-**494**

Independent atomic information in the characterization of atomic photoionization

L. A. LaJohn and R. H. Pratt

Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, Pennsylvania 15260

Abstract

Atomic photoionization at all energies can be characterized in terms of nuclear Coulomb properties (nuclear Coulombic matrix elements), plus low multipole (not beyond octupole) atomic screening contributions to the matrix elements, in obtaining predictions for differential cross sections accurate within an error limit of 1%. We indicate under what circumstances more than dipole screening contributions are needed to describe photoionization of neutral atoms. Quadrupole screening can be important except in the lightest elements. Octupole screening can be important in inner shells of intermediate Z atoms and in all shells of high-Z atoms.

PACS number(s): 32.80.Fb

I. INTRODUCTION

What atomic information is accessible in measurements of atomic photoionization? At low photon energy the process is usually described in terms of dipole matrix elements, and it is characterized in terms of a few energy-dependent parameters. In fact quadrupole effects are often not negligible. As energy increases, higher multipole contributions (characterized by additional energy dependent parameters) become important. On the other hand, as energy increases matrix elements are increasingly describable in terms of their small-distance (nuclear point Coulombic) properties. The remaining atomic information at very high energy is, for example, contained in constants such as the bound state normalization constant (i.e. the strength of the bound state wave function at the nucleus). Apart from such constants, nuclear Coulombic matrix elements (including also Coulombic phase shifts) are sufficient to accurately describe atomic photoionization at high energy [1,2], and no further atomic information is obtained in measurement of photoionization at high energies. What information is accessible at lower energies?

II. RESULTS AND DISCUSSION

We find that for low to moderate energies (below 100 keV) a description of photoionization accurate within 1% must include as many as the first three multipole screening contributions to observables U (i.e. $\Delta_j U$, as defined below, where j corresponds to the 2^j-pole screening contribution, for $j < 4$). As energy increases all Δ_j will approach zero. For total cross sections σ (i.e. integrated over angles) only dipole and quadrupole screening corrections are needed. We describe below how Δ_j depends on the n and l quantum numbers of the bound state which is photoionized, on Z (neutral atom), and on ejected photoelectron energy E. Beyond dipole Δ_j are greatest in photoionization of heavy atom inner shells, but they are significant even for outer shells and for quite light elements. While the results we present here are for independent particle approximation (IPA) matrix elements and the consequent predictions for observables, these conclusions may also be used to assess the significance of screening effects when such matrix elements are linked through electron-electron correlations [3]. Our calculations are based on our relativistic IPA code, calculating states in a potential of Dirac-Slater type, including all significant multipole and partial wave contributions.

Screening contributions enter the multipole decomposition of the total IPA photoionization matrix element M in three ways [[4]: (1) in the bound state normalization constant, i.e. the strength of the bound state wave function at the nucleus, (2) in continuum partial wave phase shifts, and (3) in reduced radial matrix elements (bound state normalization constant factored out), which determine the magnitude of the Δ_j (magnetic, unlike electric, matrix elements do not contribute significantly to Δ_j because they are very small in the energy range for which screening effects beyond bound state normalization enter the description of photoionization). The $\Delta_j U$ characterize the size of the multipole screening corrections to the physical observables U specified by the square of the total matrix element M. In this letter we will give examples of the size of non-dipole Δ_j in physical observables.

Consider physical observables $U = \sum_{l,m} U_{l,m} Y_{l.m}$ defined on a scale of unit magnitude, and the corresponding observables $U_{l,m}$ of their expansion in spherical harmonics. Such observables include angular distributions $A(E,\theta) \equiv [d\sigma/d\Omega](E,\theta)$ averaged over photon polarization and electron spin, shape functions $S(E,\theta) \equiv A(E,\theta)/\sigma(E)$ with $\sigma(E)$ the total cross section integrated over angles, and photon electron polarization correlations C_{ij}. Corresponding to U, define $U^{(j)}$ as U obtained with the reduced multipole matrix elements through order j (and all associated phase shift differences in the observable) calculated in the screened potential, the higher multipole reduced matrix elements (and associated phase shift differences) replaced by nuclear point Coulomb values. Then we define the screening contributions

$$\Delta_j U \equiv U^{(j)} - U^{(j-1)} = \sum_{lm} (\Delta_j U_{lm}) Y_{lm}(\theta,\phi) \qquad \Delta_j U_{lm} = U^{(j)}_{lm} - U^{(j-1)}_{lm}. \tag{1}$$

We now illustrate and describe more systematically the importance of the multipole screening corrections to the shape of the photoelectron angular distribution $\Delta_j S(E,\theta)$. In Fig. 1 we show the domains in (Z,n,l) (also illustrating E_b) in which dipole and quadrupole screening contributions suffice (for all energies and angles) to obtain a specified level of accuracy in the shape function. The domain in which Coulomb properties plus dipole screening contributions suffice for a 2% level of accuracy in the shape function is represented by the dark grey areas in the two panels. In the light grey areas dipole plus quadrupole $\Delta_j S$ suffice for this level of accuracy. In the white areas octupole screening contributions can exceed 2%. The next higher multipole contribution, $\Delta_4 S(E,\theta)$, never exceeds 1%; we show the boundary of its 0.3% domain. (The d-states and f-states generally show quadrupole screening contributions which exceed 2%, octupole screening corrections which can exceed 2% when $Z >\approx 50$.)

Next we discuss the magnitudes of the $\Delta_j S(E_{gm},\theta_{gm})$ (global maximum value of $\Delta_j S$, occuring at energy E_{gm} and angle θ_{gm}) as a function of (Z,n,l). In Fig. 2 we show the quadrupole and octupole magnitudes for s-state photoionization (the trends are quite similar for higher l). As Z increases, these screening contributions increase somewhat linearly. This trend is consistent with the simple analytic quadrupole retardation effects calculated by Bechler and Pratt [5] for the K and L shells (Z=6-40). (For the K-shell the Δ_2 calculated here are within about 20% of predictions from the analytic nonrelativistic formulas for all Z. The L-shell results do not agree as well.) Maximum quadrupole screening contributions are a few percent for light atoms and increase to over 40% for inner shells of heavy atoms. Octupole screening effects are smaller, less than 1% until Z exceeds 25, but can reach nearly 10% for inner shells of heavy atoms (Fig. 2). In general maximum screening effects decrease with increasing n. However they do not approach zero with increasing n, but instead saturate, in Uranium at about 10% for quadrupole contributions and 4% for octupole contributions. The trends with regard to (Z,n,l) dependence of dipole screening contributions are the opposite to those for higher multipoles. Of course the dipole effects are dominant in the case of outer shells.

Next we discuss how higher multipole screening effects (maximized for θ) depend on

E. Fig. 3 shows examples (for s-states in Uranium) of how $\Delta_2 S(E,\theta_{gm})$ and $\Delta_3 S(E,\theta_{gm})$ (global maxima for $\Delta_j S$ as a function of θ for a given E) vary with E. At low energy (less than 1 keV) $\Delta_2 S(E,\theta_{gm})$ is large for inner shells but decreases with increasing n. At energies greater than 1 keV as n increases (for constant Z) the curves for different n merge to a common curve (reflecting the fact that all ns wave functions have the same shape at the small distances which are probed at high energy), whose magnitude at fixed energy is approximately proportional to $Z^{4/3}$. (The increased oscillations for outer shells at low photoelectron energies appear to reflect the additional outer lobes of higher n states which are being probed at low energies.) For all subshells $\Delta_2 S(E,\theta_{gm})$ remains significant until near 100 keV, while at very high energy it slowly approaches zero. These features are also seen in $\Delta_3 S(E,\theta_{gm})$, of smaller magnitude, which approaches zero more rapidly with n at low energy and stays somewhat significant at higher energies. Similar behaviors are seen in all atoms, though the magnitude of higher multipole screening effects decrease at all E as Z decreases. Note that E_{gm} (the energy at which the maximum multipole screening contribution occurs) is near threshold for $n \leq 3$ but may be comparable or larger above 1 keV when $n > 3$. (It should be noted that in the full cross section, summed over multipoles, there are small non-normalization screening corrections of relativistic origin which do not go to zero in the high energy limit [6].)

Finally, we discuss the angular dependence of higher multipole screening contributions, as in shape functions and polarization correlations. Fig. 4 shows an example of the θ dependence of $\Delta_2 S(E,\theta)$ and $\Delta_3 S(E,\theta)$ for U 4s. The curves cover a wide range of photoelectron energies, showing changes in amplitude but little change of shape. This common shape, including the angles at which multipole screening effects are maximum, or near zero, is characteristic of all s-state cases, while for p states and higher the shape has some energy dependence.

We may understand these s-state features, as in the shape function, beginning from Eq. (1), which states that $\Delta_j S$ is a sum over $Y_{lm}(\theta,\phi)$ with coefficients which are proportional to $(\Delta_j S_{lm})$. [In this case only m = 0 terms are present and one usually writes $S = \sum_l B_l P_l(\theta)$].

Since higher multipole screening contributions are small in comparison to dipole terms, they enter observables such as the shape function mainly through cross terms (when present) of dipole and higher multipole matrix elements. The cross term of the dipole and jth multipole matrix elements will yield non-vanishing terms in the coefficients B_l of the shape function expansion for l through $l_{max} = j+1$ (see for example [7]); there will be non-vanishing terms for even or odd l as $j+1$ is even or odd. Knowing that the only higher multipole screening corrections which are significant are for j = 2,3 the only terms which need be considered are l = 1,3 (for j=2) and l = 2,4 (for j=3).

For s-states we can approximate the quadrupole contribution to B_1 as equal and opposite to that in B_3, likewise for the octupole contributions to B_2 and B_4. (See for example [2], which notes that for s-states the dipole amplitude is proportional to $\epsilon \cdot p$, the quadrupole amplitude to the same multiplied by $p \cdot k$, yielding a cross term, averaged over polarization, of the stated form). The s-state shapes of the quadrupole and octupole screening angular distributions are therefore (normalized to their maximum values) given by $N_{quad}(P_1 - P_3) = [(27)^{1/2}/2]x(1-x^2)$; $[N_{quad} = (27)^{1/2}/5]$ for j=2, with zeroes at x = 0,$\pm$1 (θ= 0^o, 90^o and 180^o) and extrema at x = $\pm(1/3)^{1/2}$ (θ=54.7^o and 125.3^o), corresponding to the magic angle (where dipole terms vanish, but note not octupole terms), and $N_{oct}(P_2 - P_4) = (1-5x^2)(1-x^2)$; $[N_{oct} = -8/7]$ for j=3, with zeroes at x = ±1, $\pm(1/5)^{1/2}$ ($\theta = 0^o$, 63.4^o 116.6^o and 180^o) and extrema at x = 0, $\pm(3/5)^{1/2}$ ($\theta = 39.2^o$, 90.0^o and 140.8^o) opposite in sign, the magnitude at x = 0 is $5/4^{th}$ of the other two maxima. Note both quadrupole and octupole effects vanish in this approximation at forward and backward angles; only dipole effects survive. For bound states with angular momentum $l > 0$ we can no longer approximate the quadrupole contributions to B_1 and B_3 as equal and opposite, or likewise for the octupole contributions to B_2 and B_4. As a result the positions in θ of maxima, minima, and zeros of these contributions to the shape function will be energy dependent.

We can summarize, and better understand, the importance of higher multipole screening contributions to physical observables in terms of several simple functions, for which we can offer qualitative empirical analytic expressions. We define $R(E, E_b)$ as an approximation to the ratio $R_{j+1,j}$ of reduced electric field matrix elements $M_{jl_bl_c}$ that differ by one multipole (where l_b and l_c correspond to bound and continuum state quantum numbers),

$$R_{j+1,j} = \frac{M_{j+1,l_b,l_c+1}}{M_{j,l_b,l_c}} \approx R(E, E_b) = \frac{E + (E_b/E_b + 1)^{2/3}}{E + b}, \qquad (2)$$

where b is a parameter that depends on the angular momentum l of the bound state (b=5 for $l_b = 0$ and b=20 for $l_b = 1$, taking energies in keV). The behavior of $R(E, E_b)$ as a function of E for Uranium s state photoionization is illustrated in Fig. 5a. Near threshold $R(E, E_b) \approx (1/b)[E_b/(E_b + 1)]^{2/3}$, which for large E_b approaches $1/b$ and for small E_b is proportional to $E_b^{2/3}$. When E exceeds 1 keV, $R(E, E_b)$ becomes nearly independent of E_b (or shell). The high energy maximum in higher multipole Δ_j occurs at about this energy. As E continues to increase, $R(E, E_b)$ goes to unity (at high energy all multipole matrix elements become similar in magnitude).

We define Q(E,Z) as an approximation to the ratios of screened $M^{sc}_{jl_bl_c}$ and Coulombic $M^{coul}_{jl_bl_c}$ for $j > 1$

$$Q_j(E, Z) = |1 - |\frac{M^{sc}_{jl_bl_c}}{M^{coul}_{jl_bl_c}}|| \approx Q(E, Z) = (\frac{Z^2}{aE + Z^2})^{2/3} \qquad (3)$$

using a function of E,Z having only one fit parameter a=15000 (again measuring energies in keV). $Q(E, Z)$ ranges from one (for maximum screening, at threshold) to zero for no screening at very high energy. The approach to zero occurs at a lower E for light atoms than for heavy atoms (Fig. 5b). (This expression is not appropriate for dipole $\Delta_j U(E, \theta)$, because Q_j is generally much smaller than unity near threshold, especially for inner shells.)

The general approximation for the relative magnitude of a higher multipole screening contribution may be written, in the case of an observable U such as the shape function, for which higher multipole matrix elements enter linearly in interference terms,

$$\Delta_j U(E, E_b, Z, \theta) = [R(E, E_b)]^{j-1} Q(E, Z) T(\theta). \qquad (4)$$

The function T reflects the angular dependence of the screening contributions; the s-state form is being used to approximate for all subshells. The first two terms in (4) are due to the reduced electric field matrix element and determine the magnitude of the screening contribution. Screening effects also depend on θ [$T(\theta)$) in equation (4)], examples of this will be given. The product of $R(E, E_b)$ and $Q(E, Z)$ (for j=2) as a function of E is given in Fig. 5c. The general behavior of $\Delta_2 S(E, \theta_{gm})$ is produced. The threshold amplitude becomes very small for low binding energy. Similar results can be obtained for octupole screening effects. In the total reduced cross section, higher multipole contributions would enter quadratically, corresponding to squaring (4) with $T = 1$. If for E near E_{gm} Q is near 1, we can expect the magnitude of quadrupole screening effects in total cross sections to correspond to the magnitude of octupole screening effects in angular distributions.

We should caution that we have only estimated over-all magnitudes. For example, we have not considered the phase shift differences which multiply reduced matrix elements. These can modify the size of $\Delta_j U$ at specific energies, causing oscillations with energy, but will not greatly affect the magnitudes more globally. Specific calculations are required for a comparison with any given specific experiment; what we have given here are cruder estimates which can focus attention on possible parameter domains which merit examination.

III. SUMMARY

In summary, our results demonstrate that at all energies atomic photoionization can be characterized in terms of a very limited number of measurable atomic parameters associated with independent particle approximation matrix elements. At high energy only Coulomb matrix elements are needed, while at lower energies one must include low multipole screening contributions, but never higher than octupole, in order to make predictions accurate within 1%. Quadrupole screening can be important except in the lightest elements. Octupole screening can be important in inner shells of intermediate Z atoms and in all shells of high-Z atoms. These results should be relevant to experimental measurements of nondipole effects in photoionization angular distributions [8].

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation under Grant No. PHY-9601752.

IV. FIGURES

REFERENCES

[1] R. H. Pratt, A. Ron and H. K. Tseng, Rev. Mod. Phys. **45**, 273 (1973).

[2] H. K. Tseng, R. H. Pratt, S. Yu and A. Ron, Phys. Rev. A **17**, 1061 (1978).

[3] E. W. B. Dias, H. S. Chakraboty, P. C. Deshmukh, S. T. Manson, O. Hemmers, P. Glans, D. L. Hanson, H. Wang, S. B. Whitfield, D. W. Lindle, R. Wehlitz, J. C. Levin, I. A. Sellin, R. C. C. Perera, Phys. Rev. Lett, **78**, 4553 (1997).

[4] R. H. Pratt and L. LaJohn, Nucl. Instrum. Methods Phys. Res. B, **99**, 136 (1995).

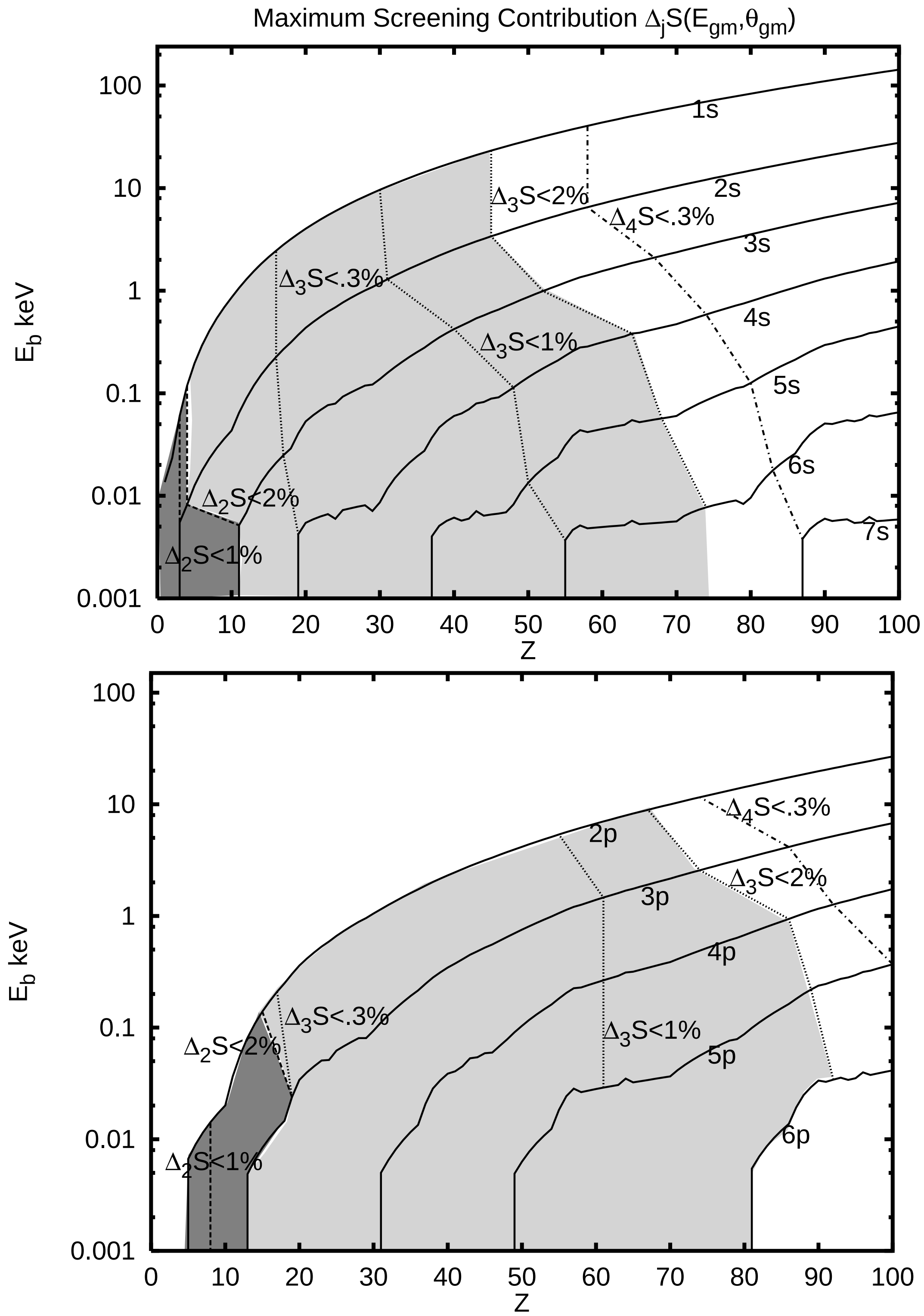
[5] A. Bechler and R. H. Pratt, Phys. Rev. A **39**, 1774 (1989); **42**, 6400 (1990).

[6] R. Y. Yin, O. V. Gabriel and R. H. Pratt, Phys. Rev. A **36**, 1207 (1987).

[7] K. N. Huang, Phys. Rev. A **22**, 223 (1980); **26**, 3676 (1982).

[8] M. Jung, B. Krassig, D. S. Gemmell, E. P. Kanter, T. LeBrun, S. H. Southworth, and L. Young, Phys. Rev. Lett. **54**, 2127 (1996).

FIGURES



Maximum Screening Contribution $\Delta_j S(E_{gm},\theta_{gm})$
E_b keV
Z
1s
2s
3s
4s
5s
6s
7s
$\Delta_3S<2\%$
$\Delta_4S<.3\%$
$\Delta_3S<.3\%$
$\Delta_3S<1\%$
$\Delta_2S<2\%$
$\Delta_2S<1\%$
2p
3p
4p
5p
6p

FIG. 1. Domain in (Z,n,l) in which $\Delta_j S(E,\theta)$ for low multipoles are sufficient to achieve a given level of accuracy. The solid curves correspond to binding energies for $s_{1/2}$ and $p_{1/2}$ states (upper and lower graphs respectively) for Z ranging from 1 to 100. The dashed lines correspond to the boundary at which quadrupole screening contributions can reach 1% (leftmost line) and 2% (boundary for dark shaded area, within which dipole screening is sufficient for 2% accuracy). The dotted lines correspond to the 0.3%, 1% and 2% limits for octupole screening contributions (the latter is the boundary for the lightly shaded area, within which screening corrections through quadrupole are sufficient for 2% accuracy). In the white area octupole screening contributions to $S(E,\theta)$ can exceed 2%. The dot-dashed line is the 0.3% boundary for 2^4-pole screening contributions. We have omitted $p_{3/2}$ results because they would be almost the same as the $p_{1/2}$ values.

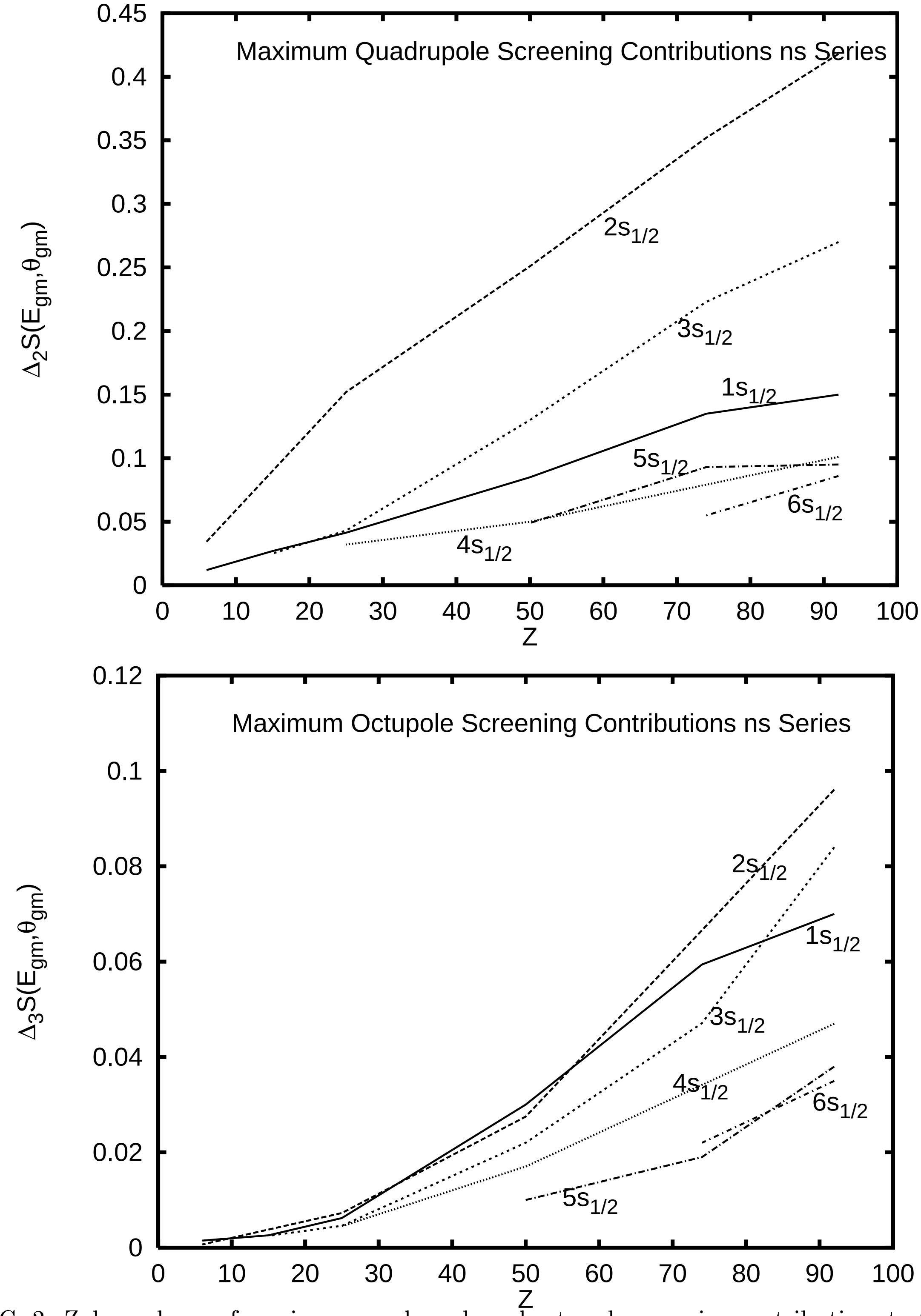


FIG. 2. Z dependence of maximum quadrupole and octupole screening contributions to $S(E,\theta)$ for $s_{1/2}$ states [i.e. $\Delta_j S(E_{gm},\theta_{gm})$, j=2, 3].

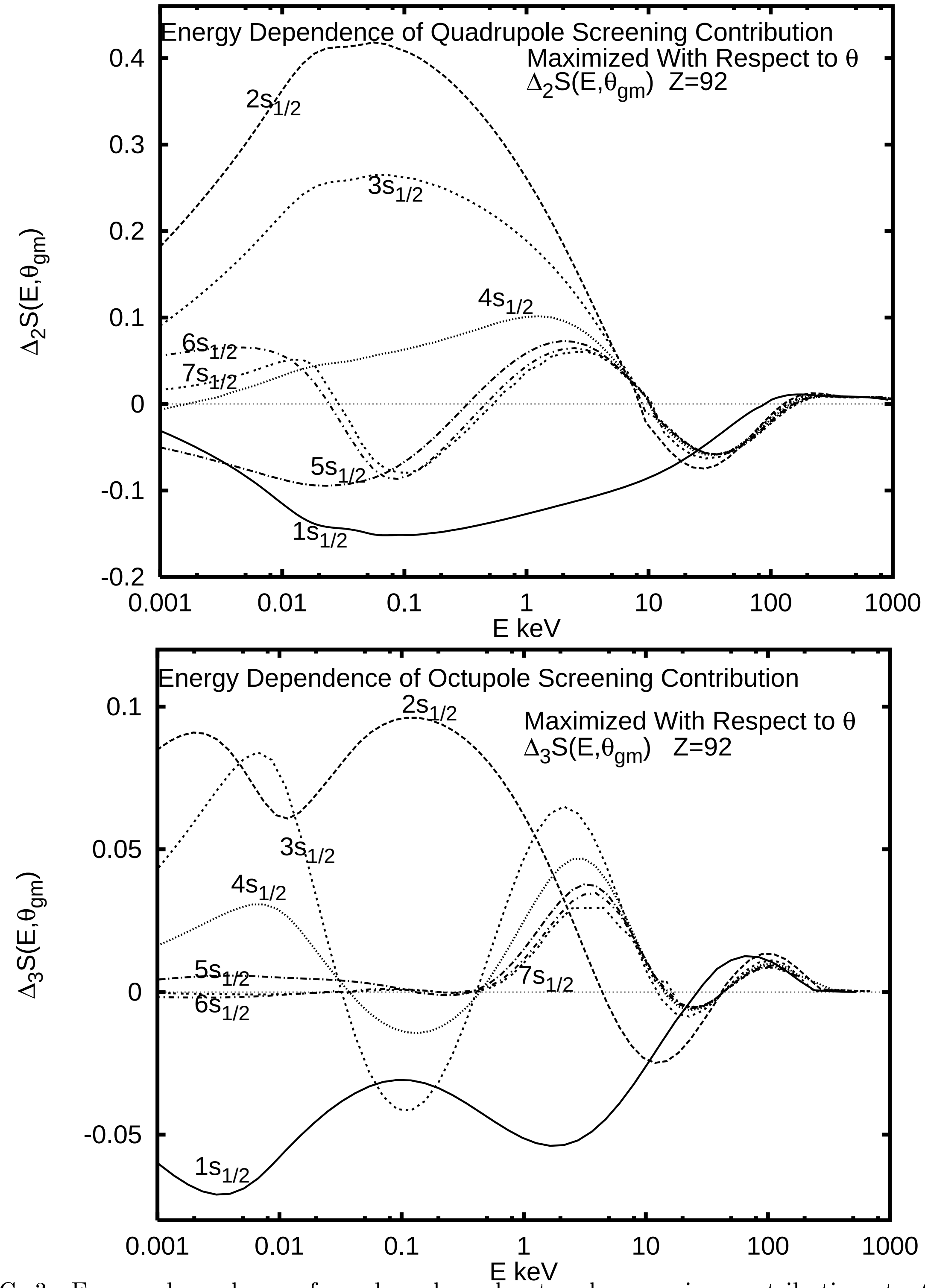


FIG. 3. Energy dependence of quadrupole and octupole screening contributions to $S(E,\theta)$ (upper and lower graphs respectively) for Uranium $s_{1/2}$ state photoionization, maximized with respect to the electron emission angle [i.e. $\Delta_j S(E,\theta_m)$].

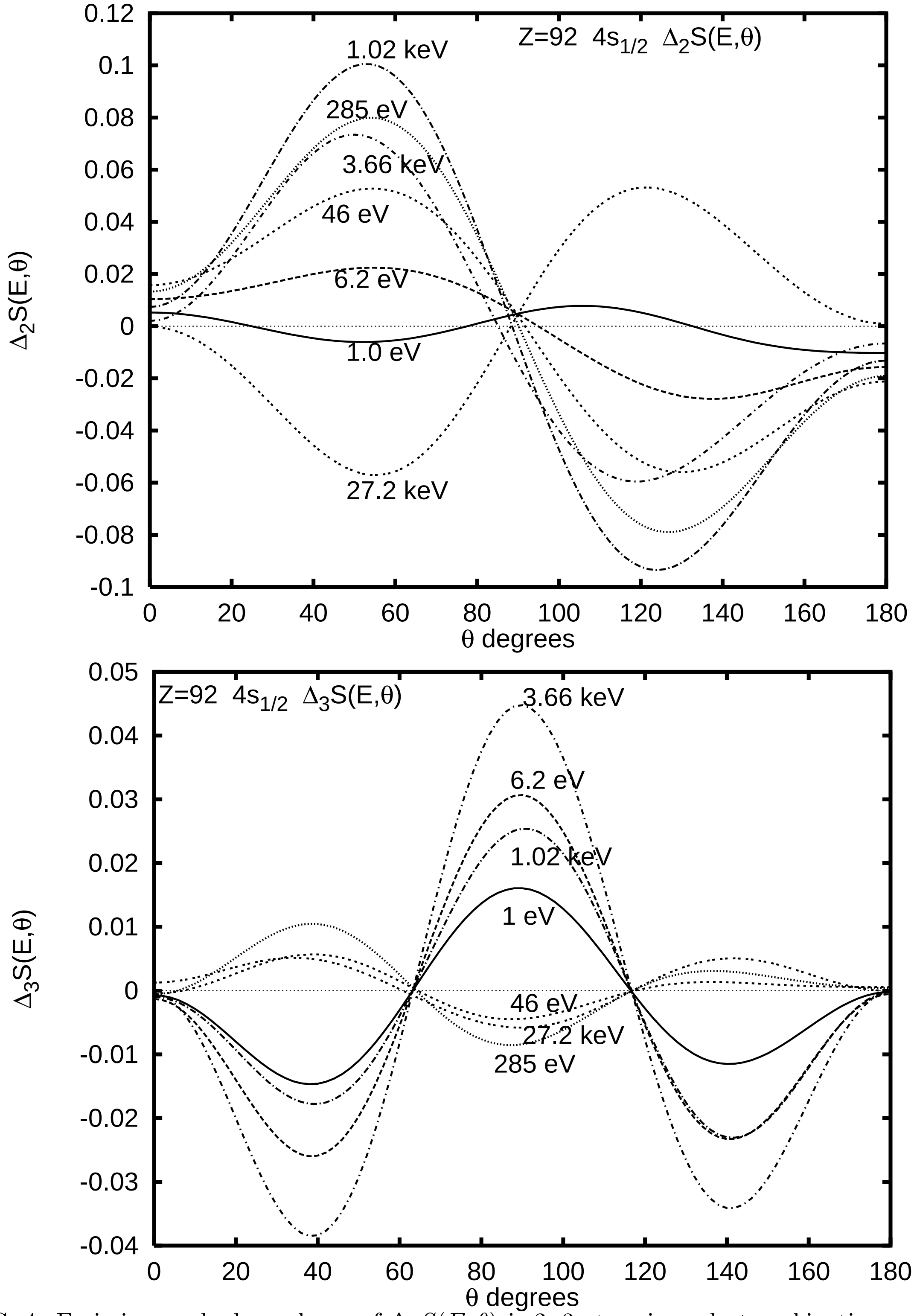
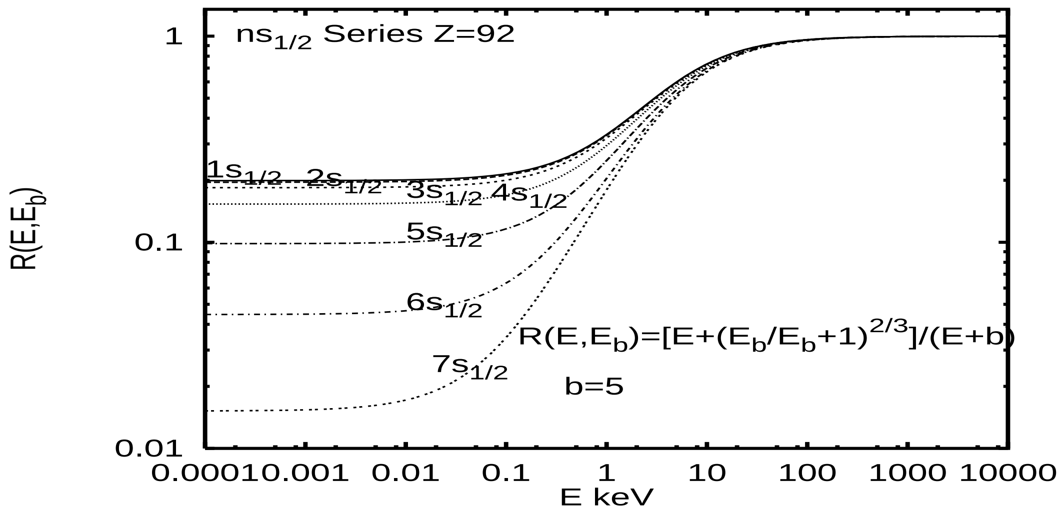
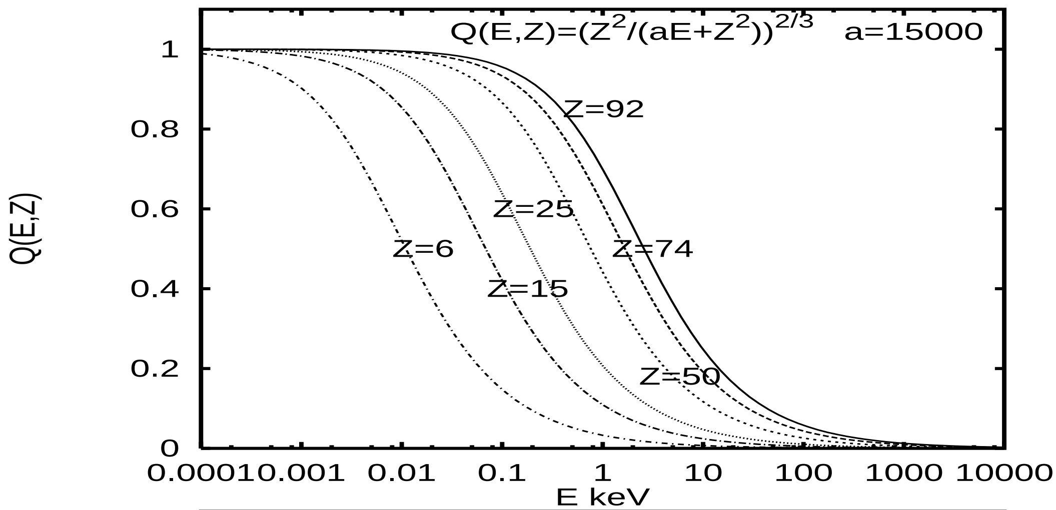


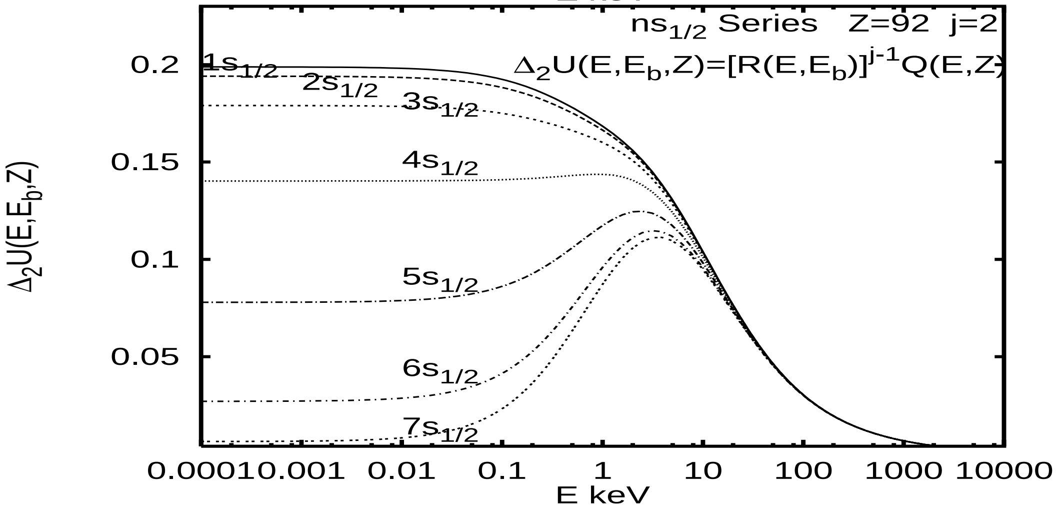
FIG. 4. Emission angle dependence of $\Delta_j S(E,\theta)$ j=2, 3 at various electron kinetic energies in the case of Uranium $4s_{1/2}$ photoionization.



ns1/2 Series Z=92
1s1/2
2s1/2
3s1/2
4s1/2
5s1/2
6s1/2
7s1/2
R(E,Eb)=[E+(Eb/Eb+1)2/3]/(E+b)
b=5
R(E,Eb)
1
0.1
0.01
0.00010.001 0.01 0.1 1 10 100 1000 10000
E keV



Q(E,Z)=(Z2/(aE+Z2))2/3 a=15000
Z=92
Z=25
Z=6
Z=74
Z=15
Z=50
Q(E,Z)
1
0.8
0.6
0.4
0.2
0
0.00010.001 0.01 0.1 1 10 100 1000 10000
E keV



ns1/2 Series Z=92 j=2
Δ2U(E,Eb,Z)=[R(E,Eb)]j-1Q(E,Z)
1s1/2
2s1/2
3s1/2
4s1/2
5s1/2
6s1/2
7s1/2
Δ2U(E,Eb,Z)
0.2
0.15
0.1
0.05
0.00010.001 0.01 0.1 1 10 100 1000 10000
E keV

FIG. 5. For Uranium $s_{1/2}$ states, (a) estimated photoelectron energy E dependence of the ratio of reduced electric field matrix elements that differ by one multipole, $R(E, E_b)$ [see equation (2)], (b) estimated E dependen of the ratio of screened to Coulombic reduced electric field matrix elements $Q(E, Z)$ [see equation (3)], (c) estimated E dependence of quadrupole screening contribution to physical observable U [see equation (4)].