Dependence of atomic photoeffect on a bound-electron magnetic substate*

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Flügge, Mehlhorn, and Schmidt have suggested that the dependence of the atomic photoeffect on a bound-electron magnetic substate will have observable consequences for the angular distribution of subsequent Auger electrons. Because of the constraints of parity and time-reversal invariance, this possibility first arises for ejection from $L_{\rm III}$ and $M_{\rm III}$ subshells. We have calculated the relative probabilities of $|j_z| = 3/2$ and $|j_z| = 1/2$ ejection from these subshells, both with the relativistic Born approximation (Gavrila's formulation) and in the nonrelativistic dipole approximation. The ratio of these cross sections can have a dramatic energy dependence, ranging from 7 at high energies to 9/11 in the low-energy domain, and dropping to 1/3 near threshold if there is a Cooper minimum. Screening effects are discussed.

I. INTRODUCTION

In most work on photoabsorption it has been assumed that the magnetic substate from which an electron is ejected is not observed. Consequently, a sum has been performed over quantum numbers j_z corresponding to a given subshell. Extensive discussions and calculations of the resulting total photoelectric cross sections are now available.[1-4]

However, it has been suggested that the dependence of the photoeffect on magnetic substate, although not directly measured, could be determined from observation of the transition which fills the resultant vacancy, either through transition radiation or Auger emission.[5] Cooper and Zare[6] argued that Auger electron angular distributions would be isotropic and would not provide information on magnetic substates. However, more recently Flügge, Mehlhorn, and Schmidt[7] (FMS) exhibited significant anisotropy in explicit calculations and concluded that the magnetic-substate dependence of total photoelectric cross sections can be determined experimentally.[8] We agree with these results of FMS and wish to emphasize that the difference from the Cooper-Zare conclusion is independent of the choice of representation.[9]

If experimental interest warrants, it would be possible to redo the "exact" numerical photoeffect calculations and obtain quantitative predictions for the magnetic-substate dependence with an accuracy comparable to that achieved for other predictions for a given energy, shell, and target element Z. But meanwhile, to obtain some general insight, it seems useful to discuss what can be said about magnetic-substate dependence using analytic methods. In this paper we will use the relativistic Born approximation and nonrelativistic dipole approximation, together with a discussion of screening effects, to indicate how magnetic-substate dependence varies with energy and with shell and subshell.

We begin in Sec. II with a brief description of the formalism for these calculations. We show that parity and time-reversal invariance imply cross sections from the two substates $\pm j_z$ are the same, so that interesting effects first arise in the $L_{\rm III}$ subshell. FMS note that the conclusion of Cooper and Zare was obtained in an (l, m_l) representation. We will show that the result of FMS can be obtained in either representation. We comment on situations in which interference terms may be neglected and for which it is adequate to assume we are observing photoelectric cross sections (rather than considering matrix elements). In such cases the relative cross sections from different magnetic substates will give the probabilities of the initial state for the subsequent transition process. In Sec. III we generalize Gavrila's[10] relativistic Born-approximation calculation for the $L_{\rm III}$-subshell photoeffect to include magnetic-substate dependence, finding a dramatic energy dependence. Using photoeffect normalization theory[11] these results may also be applied to higher $P_{3/2}$ subshells. We discuss the range of validity of such results and how they are modified by screening. In Sec. IV we present two types of nonrelativistic dipole-approximation results. We give analytic point-Coulomb predictions for arbitrary shell and subshell, discuss when they can be used and how screening should be treated, and show the connection with the relativistic results in the $P_{3/2}$ case. We also present, for comparison, some L- and M-shell numerical results in screened potentials, which help to delineate the situations where analytic results can be useful. The dramatic variation at low energies for $M_{\rm III}$ found by FMS, using McGuire's data,[12] is a simple consequence of the presence of a Cooper minimum.[13,14]

II. FORMULATION

The atomic photoelectric effect occurs if the energy of an incident photon k is greater than the binding energy E_b of an electron in the atom and the electron is raised into a continuum state. In the case of inner-shell ionization the vacancy can be filled in an Auger transition (or a radiative transition). We may write the matrix elements for these processes as

$$\begin{aligned} M_{\text{photo}} &= \langle N, p | M | I \rangle , \\ M_{\text{Auger}} &= \langle F, p, p' | A | N, p \rangle , \\ M_{\text{rad}} &= \langle F', p | R | N, p \rangle . \end{aligned} \tag{1}$$

Here I is the initial state of the atom and N the intermediate (once-ionized state) of the atom; F and F' are final (twice- and once-ionized) states of the atom; p is the momentum of the photoelectron and p' the momentum of the Auger electron.

The differential cross section for the atomic photoeffect, averaged over initial photon polarization ϵ, is

$$\frac{d\sigma}{d\Omega} = \frac{(2\pi)^2 e^2 \hbar^3}{m^2 ck} \sum_{\epsilon} |M_{\text{photo}}|^2 \tag{2}$$

subject to energy conservation; the total cross section is obtained by integrating over the final photoelectron variables. However, if the intermediate state N is not observed, but rather the final state F (or F'), through detection of the Auger electron p' (or the transition radiation), then following FMS the observed cross sections correspond to

$$\begin{aligned} &\text{const} \times \sum_{\epsilon} \Big| \sum_{N} \langle F, p, p' | A | N, p \rangle \langle N, p | M | I \rangle \Big|^2 , \\ &\text{const} \times \sum_{\epsilon} \Big| \sum_{N} \langle F', p | R | N, p \rangle \langle N, p | M | I \rangle \Big|^2 , \end{aligned} \tag{3}$$

in the two cases.[15] In the situation discussed by FMS it is assumed that we do not observe (i.e., sum over) the photoelectron p, so that we wish to integrate (3) over photoelectron variables. As long as we assume randomly oriented initial atoms and unpolarized radiation, (3) then reduces to a product of cross sections,

$$\begin{aligned} &\text{const} \times \sum_{J_z^N} |\langle F, p' | A | N(E_b, J_z^N) \rangle|^2 \bar{\sigma}(E_b, J_z^N, k) , \\ &\text{const} \times \sum_{J_z^N} |\langle F' | R | N(E_b, J_z^N) \rangle|^2 \bar{\sigma}(E_b, J_z^N, k) , \end{aligned} \tag{4}$$

where $\bar{\sigma}$ is the photoelectric cross section corresponding to ejection of a photoelectron of energy $k - E_b$, leaving the atom in the magnetic substate J_z^N, averaged over initial orientation and photon polarization. The Z axis has been taken along $\hat{k}$. This follows because if we are performing these sums and averages we can take the photoelectron variables as j^p, j_z^p (rather than momentum) and quantize photon helicity λ and atomic orientation J^I along the z axis. Then for given j_z^p, λ, J_z^I the magnetic substate of N is determined as $\lambda + J_z^I - j_z^p \equiv J_z^N$. The total energy of the state N differs from that of I by E_b. Assuming that total energy and magnetic substate completely characterize an atomic state, the sum over N in Eq. (3) reduces to a single term. Then the sums over λ, J_z^I, and j_z^p can be replaced by sums over λ, J_z^I, and J_z^N. The Auger and radiative matrix elements do not depend on λ and J_z^I, since they do not depend on the photoelectron p but only on N, and hence these sums act only on the photoelectric cross section, leading to Eq. (4).

We have used the (J, J_z) rather than an (L, M_L) representation above, but it is clear that the argument can be carried through in either case. In fact we find M_L^N dependence in $\bar{\sigma}(E_b, M_L^N, k)$ in model calculations, contrary to Cooper and Zare. Note, however, that the amount of anisotropy will depend on the experimental situation, i.e., whether transitions from L_{III} states are distinguished from L_{II} cases. Cooper and Zare's conclusion appears to simply be a misinterpretation of their formulas, which are correct.[9]

We will restrict our subsequent discussion to a simple atomic model, in which all electrons move in a common central potential. Exchange and correlation effects are neglected, and the matrix elements (1) can be evaluated in terms of single-particle electron wave functions. (The results presented by FMS are of this type.) Then, in a relativistic formulation,

$$M_{\text{photo}} = \int d^3r\, \psi_f^\dagger \vec{\alpha} \cdot \vec{\epsilon} e^{i\vec{k}\cdot\vec{r}} \psi_i , \tag{5}$$

where ψ_i is the initial bound state, from which an electron is removed, ψ_f is the final photoelectron, and $(\vec{k}, \vec{\epsilon})$ are the momentum and polarization of the initial photon. In the nonrelativistic case $\vec{\alpha}$ is replaced by $\vec{p}$.

In this single-electron model it is easy to prove that the photoeffect total cross section, averaged over photon polarization and summed over photoelectron spins, is the same for the ejection of an electron from the magnetic substates $\pm j_z$. First we show that the total cross section $\sigma(\lambda, j_z^p, j_z)$ for ejection of a photoelectron of definite j and j_z^p by a photon of helicity λ is the same as the cross section $\sigma(-\lambda, -j_z^p, -j_z)$, all other quantum numbers remaining unchanged. Using parity and time-reversal invariance on electron states of definite j, up to a phase,

$$M^*_{\text{photo}}(\lambda, j^p_z, j_z) = \int d^3r\, (PT\psi^*_f)^\dagger \times T\vec{\alpha}^*\cdot\vec{\epsilon}^*T^{-1}(PT\psi^*_i)e^{+i\vec{k}\cdot\vec{r}} = M_{\text{photo}}(-\lambda, -j^p_z, -j_z), \quad (6)$$

where $j^p_z = \lambda + j_z$. This follows because[2] $PT\psi^*$ is also a solution of the Dirac equation of the same energy and angular momentum, but opposite magnetic substate, and because $T\vec{\alpha}^*\cdot\vec{\epsilon}^*T^{-1} \equiv \vec{\alpha}\cdot\vec{\epsilon}^*$ if $\vec{\epsilon}$ specifies a definite helicity (and then $\vec{\epsilon}^*$ is of opposite helicity). Hence the corresponding total cross sections are equal. Finally, summing over λ and j we obtain the asserted result. Note that we needed unpolarized photons and a sum over photoelectron spins in order to have the freedom to pick helicity states and states of definite j^p_z for our complete set. It is clear that a corresponding nonrelativistic result for $\pm j_z$ or $\pm m_l$ can also be proved. These results have the consequence that nontrivial magnetic-substate dependence is only possible for levels with $j \geq \frac{3}{2}$ or $l \geq 1$.

III. RELATIVISTIC BORN APPROXIMATION

At high energies the relativistic Born approximation in a point-Coulomb potential may be used to estimate the magnetic-substate dependence of the photoeffect. We use the momentum space formalism of Gavrila,[10] in which the matrix element for the photoeffect becomes

$$M_{\text{photo}} = \int \psi^\dagger_f(\vec{r})\vec{\alpha}\cdot\vec{\epsilon}\, e^{i\vec{k}\cdot\vec{r}}\psi_i(\vec{r})d^3r = \int u^*_f(\vec{q})\vec{\alpha}\cdot\vec{\epsilon}u_i(\vec{q}-\vec{k})d^3q\,. \quad (7)$$

The $u(\vec{q})$ are the momentum-space wave functions. As is well known, one must expand the continuum state to second order in $Z\alpha$ to obtain the photoeffect matrix element correct to first order in $Z\alpha$. The relativistic energy-conservation relation may be taken as $E = (p^2+1)^{1/2} = k+1$ in the units $m = \hbar = c = 1$ used in this section, as the binding energy $O((Z\alpha)^2)$ can be neglected in this approximation.

The continuum wave function $u_f(\vec{q})$ satisfies the Dirac equation in momentum space:

$$(\not{q} - 1)u_f(\vec{q}) = ie\int \not{A}(\vec{l}-\vec{q})u_f(\vec{q})d^3q\,, \quad (8)$$

where q is the four-vector $(E, \vec{q})$ and $2\pi^2 A(\vec{l}) = -Z\alpha/(l^2+\mu^2)$. The Born-approximation expansion for this wave function begins with the terms

$$u_f(\vec{q}) = (pE)^{1/2}\left(\delta(\vec{q}-\vec{p}) - ie\frac{\not{q}+1}{\vec{q}^2-\vec{p}^2-i\epsilon} \times \not{A}(\vec{p}-\vec{q}) + \dots\right)\chi_f(\vec{p}), \quad (9)$$

where $\chi_f(\vec{p}) = (1,0,0,0)^\dagger$ for spin-up and $(0,1,0,0)^\dagger$ for spin-down.

Following Gavrila, the L_{III} wave function may be written

$$u_i(\vec{q}) = \frac{1}{(4\pi)^{1/2}}\left(G(\vec{q}) + F(\vec{q})\gamma_0\frac{\vec{\gamma}\cdot\vec{q}}{q}\right)\left(Z^{1,3/2,j_z}_{0}(\vec{q}/q)\right), \quad (10)$$

with

$$G(\vec{q}) = \frac{2}{(3\pi)^{1/2}}(\alpha Z)^{7/2}\frac{q}{(\vec{q}^2+\frac{1}{4}\alpha^2Z^2)^3} \quad F(\vec{q}) = \tfrac{1}{2}qG(\vec{q});$$

$Z_{1,3/2,j_z}$ is a Pauli spinor.[16] Terms of $u_i(\vec{q})$ which do not contribute to lowest order in $Z\alpha$ in the matrix element have been omitted. Equation (10) may be rewritten in the more convenient form

$$u_i(\vec{q}) = (2/3\pi)^{1/2}(\alpha Z)^{7/2}\frac{1}{(\vec{q}^2+\frac{1}{4}\alpha^2Z^2)^3} \times (1+\tfrac{1}{2}\gamma_0\vec{\gamma}\cdot\vec{q})(\gamma'\cdot\vec{q})\gamma_0\gamma_5\chi_i\,, \quad (11)$$

where

$$\gamma' = (-\gamma_1, -\gamma_2, 2\gamma_3) \quad \text{for } j_z = \pm\tfrac{1}{2}$$
$$= ((3)^{1/2}\gamma_1, -(3)^{1/2}\gamma_2, 0) \quad \text{for } j_z = \pm\tfrac{3}{2}.$$

Again $\chi_i = (1,0,0,0)^\dagger$ for positive j_z and $(0,1,0,0)^\dagger$ for negative j_z. Using Eqs. (7) and (11) we can express the matrix element in the form $M_{\text{photo}} = \bar{\chi}_f Q\chi_i$. The matrix Q depends on the magnitude of j_z; the spinor χ_i on the sign of j_z. The differential cross section summed over final electron-spin state f is

$$\frac{d\sigma}{d\Omega} = (2\pi)^2\frac{\alpha}{k}\frac{1}{2}\sum_{\epsilon,f}|M_{\text{photo}}|^2\,. \quad (12)$$

After a very complicated trace calculation, followed by an integration over photoelectron directions, we obtain the total cross sections summed over final spins.[17] In agreement with Sec. II the results are independent of the sign of j_z. We find

$$\sigma(j_z = \pm\tfrac{3}{2}) = G\left(\frac{E^3}{2} + \frac{3E^2}{10} + \frac{17E}{20} + \frac{21}{20} - \frac{4E^2 - 12E + 13 - 3/(E-1)}{4(E^2-1)^{1/2}} \times \ln[E + (E^2-1)^{1/2}]\right)$$

and

$$\sigma(j_z=\pm\tfrac{1}{2})=G\left(\frac{7E^3}{2}-\frac{63E^2}{10}+\frac{83}{20}E+\frac{39}{20}-\frac{3+3/(E-1)}{4(E^2-1)^{1/2}}\times\ln[E+(E^2-1)^{1/2}]\right), \tag{13}$$

with

$$G=(\pi/24)\alpha^8 Z^7[(E^2-1)^{1/2}/(E-1)^5].$$

We shall now discuss the expected validity of these results, which assume the relativistic Born approximation and neglect screening. The relativistic Born approximation is expected to be valid when $Z\alpha/\beta \ll 1$. Thus, at best (i.e., for high energies) it is good for $Z\alpha<0.2$. For lower energies, the method remains valid for the lightest elements. (Since we are concerned with a ratio of cross sections which to lowest order is independent of $Z\alpha$ it is possible that, like other photoeffect polarization correlations independent of $Z\alpha$ in lowest order, the Born series here actually converges somewhat better.) For Z such that $Z\alpha<0.2$ there is a range of energies for which the relativistic Born condition $Z\alpha/\beta\ll 1$ and the nonrelativistic condition $\frac{1}{2}p^2\ll 1$ can both be approximately satisfied. Hence we expect relativistic Born predictions to agree with nonrelativistic results (not Born approximation) in such ranges. The normalization screening theory notes that the shapes of electron wave functions are independent of energy and of screening near the origin (electron Compton wavelength distances). When the relativistic Born approximation is valid the important regions of the photoeffect matrix element are at small distances (except in the lightest elements $Z<4$) and the effect of screening arises from the normalization of the bound-state wave function. (The normalization of a continuum wave function of such energy is nearly independent of screening.) In the ratio of cross sections being calculated here the bound-state normalization cancels out and so screening does not affect the ratio obtained in the relativistic Born approximation. Further, since the wave-function shapes are independent of energy, the ratio of cross sections obtained from Eq. (13) applies not just to the L_{III} shell but also to M_{III} and all the $P_{3/2}$ shells. We show in Fig. 1 the predictions of Eq. (13) for the ratios of cross sections. The curves above 20 keV were obtained with the relativistic Born approximation. We also note two limiting cases, nonrelativistic (NR) ($E\to 1$) and extreme relativistic (ER) ($E\to\infty$):

$$\lim_{E\to 1}\frac{\sigma(P_{3/2},j_z=\pm\frac{1}{2})}{\sigma(P_{3/2},j_z=\pm\frac{3}{2})}=\frac{9}{11},\quad \text{NR}$$

$$\lim_{E\to\infty}\frac{\sigma(P_{3/2},j_z=\pm\frac{1}{2})}{\sigma(P_{3/2},j_z=\pm\frac{3}{2})}=7,\quad \text{ER}. \tag{14}$$

The energy dependence of the ratio of cross sections for the two magnetic substates is thus very dramatic in the relativistic region. The $j_z=\pm\frac{1}{2}$ electrons are much more likely to be ejected at higher energies, and $j_z=\pm\frac{3}{2}$ electrons at lower energies.

IV. NONRELATIVISTIC DIPOLE APPROXIMATION

If the incident photon energy is small compared to the electron mass mc^2, then the energy of the photoelectron is also small compared to mc^2 and nonrelativistic approximations may apply. If the more stringent condition $h\nu \ll Z\alpha mc^2$ is also satisfied, then the photon wavelength is also large compared to the radius of the bound electron. In this case we can neglect retardation and use the dipole

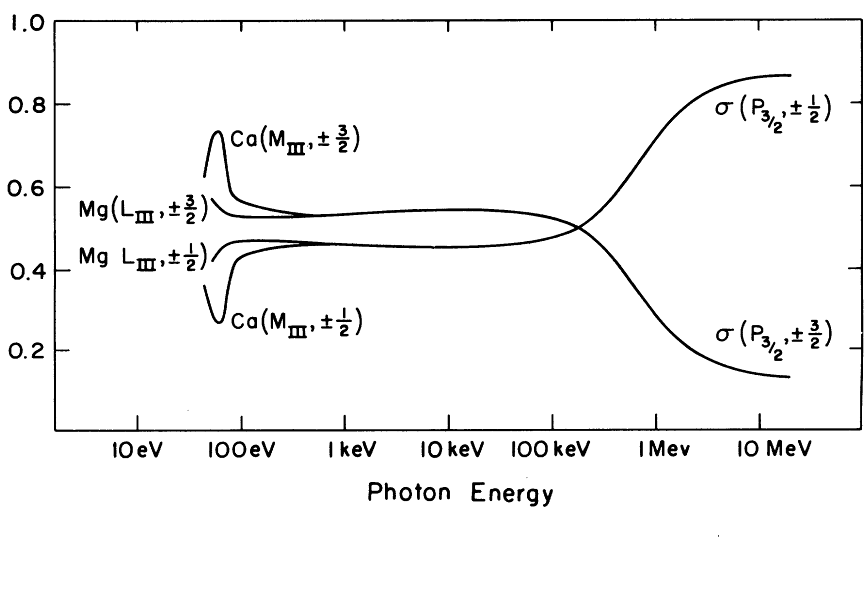

FIG. 1. Fractions of $P_{3/2}$ total photoelectric cross sections corresponding to ejection of $|j_z|=\frac{3}{2}$ and $|j_z|=\frac{1}{2}$ electrons, for Ca and Mg and photon energies from threshold to 10 MeV. Predictions above 20 keV were obtained with relativistic point-Coulomb–Born approximation; predictions below 20 keV were obtained with a nonrelativistic dipole calculation in a screened potential.

approximation. In fact one finds that, when the higher multipole terms become important in the total cross section, so do relativistic effects.[18] So no useful purpose is served by including retardation in our calculation of the nonrelativistic matrix element. Here we will discuss analytic point-Coulomb and numerical screened calculations in nonrelativistic dipole approximation.

The cross section in the dipole approximation is

$$\propto \sum_{l^p m_l^p, m_s^p} |M^{\lambda}_{\rm photo}(j,j_z)|^2 \propto \sum_{l^p, m_l^p, m_s^p} |\langle l^p, s^p, m_l^p, m_s^p | r Y_{1,\lambda} | l, s, j, j_z\rangle|^2 . \qquad (15)$$

Here λ specifies the helicity of the photon and $|l^p, s^p, m_l^p, m_s^p\rangle$ are the possible continuum states in a dipole transition. For an unpolarized incident beam we obtain a total cross section

$$\propto \sum_{\lambda} \int d\Omega_p \sum_{l^p, m_l^p, m_s^p} |M^{\lambda}_{\rm photo}(j,j_z)|^2 \propto \sum_{m_l, m_s} \begin{pmatrix} l & s & j \\ m_l & m_s & j_z \end{pmatrix}^2 \frac{l(l-1)+m_l^2}{2l-1} |R_{l-1}|^2 + \frac{(l+1)(l+2)+m_l^2}{2l+3} |R_{l+1}|^2 , \qquad (16)$$

where

$$R_{l\pm 1} = \int_0^\infty R^*_{k,l\pm 1} r^3 R_{nl}\, dr .$$

Here $R_{k,l\pm1}$ and R_{nl} are continuum and bound radial wave functions.

Then the ratio of the cross sections for $P_{3/2}$ is

$$\frac{\sigma(P_{3/2}, j_z = \pm\frac{1}{2})}{\sigma(P_{3/2}, j_z = \pm\frac{3}{2})} = \frac{|R_{l-1}|^2 + \frac{19}{5}|R_{l+1}|^2}{3|R_{l-1}|^2 + \frac{21}{5}|R_{l+1}|^2} . \qquad (17)$$

If we calculate $R_{l\pm1}$ in Born approximation, we find that

$$R_{l+1}/R_{l-1} = 2l . \qquad (18)$$

From our previous discussion, we expect this result to be independent of screening when the Born approximation is valid ($\eta \equiv Z\alpha/p \ll 1$). From Eqs. (17) and (18) we have

$$\frac{\sigma(P_{3/2}, j_z = \pm\frac{1}{2})}{\sigma(P_{3/2}, j_z = \pm\frac{3}{2})} = \frac{9}{11} ,$$

in agreement with the nonrelativistic limit of the relativistic Born approximation. The ratios of the dipole radial matrix elements can be calculated explicitly for the point-Coulomb potential without using the Born approximation by the factorization method.[19] The needed recurrence relations are

$$2lA^l_n R^{l-1,l}_{n,l\eta} = (2l+1) A^{l+1}_{l\eta} R^{l,l+1}_{n,l\eta} + A^{l+1}_n R^{l+1,l}_{n,l\eta} ,$$
$$2lA^l_{l\eta} R^{l,l-1}_{n,l\eta} = A^{l+1}_{l\eta} R^{l,l+1}_{n,l\eta} + (2l+1) A^{l+1}_n R^{l+1,l}_{n,l\eta} , \qquad (19)$$

with

$$A^l_n = (n^2 - l^2)^{1/2}/nl , \quad A^l_{l\eta} = (\eta^2 + l^2)^{1/2}/\eta l$$

and

$$R^{l,l'}_{n,l\eta} = \int R^{l'*}_{l\eta} r^3 R^l_n d^3 r .$$

Simple calculation gives the ratios:

$$\frac{R_{l+1}}{R_{l-1}} = 2l \left(\frac{\eta^2 + l^2}{\eta^2 + (l+1)^2}\right)^{1/2} \frac{l+1}{l} \quad \text{for } n = l+1$$
$$= 2l \frac{\{(\eta^2 + l^2)[\eta^2 + (l+1)^2]\}^{1/2}}{l(l+1) + \eta^2 l(l+5)/(l+2)^2} \quad \text{for } n = l+2 . \qquad (20)$$

The ratio of the cross sections from the magnetic subshells for the case of $n = l+1$ and $l+2$ is then

$$\frac{\sigma(j_z = \pm\frac{1}{2})}{\sigma(j_z = \pm\frac{3}{2})} = \frac{103\eta^2 + 108}{117\eta^2 + 132} \quad \text{for } L_{\rm III}$$
$$= \frac{176\eta^4 + 885\eta^2 + 729}{204\eta^4 + 1035\eta^2 + 891} \quad \text{for } M_{\rm III} . \qquad (21)$$

The value of $\frac{9}{11}$ (for $\eta \sim 0$) agrees with the Born-approximation result, confirming our earlier assertion that there was a region of overlap between the Born approximation and nonrelativistic dipole approximation. We have indicated that in this region screening may be neglected. As η increases the energy is decreasing and we may expect the important regions of the matrix elements to be larger, perhaps characterized by an effective charge $Z_{\rm eff}$ which changes the value of η. Since the ratio is not sensitive to η this suggests that the ratio will not be sensitive to screening. This agrees with numerical data of McGuire[12] and Kissel[20] at energies of 2–10 times binding energies, as shown in Fig. 1.

At lower energy, very near the threshold, the outer region begins to dominate in the radial integration ($1/r$ potential). At a certain energy R_{l+1} vanishes and then changes sign for $M_{\rm III}$ levels. This phenomena, called a Cooper minimum, can be explained by quantum defect theory. At a Cooper minimum, we see from Eq. (17) that $\sigma(j_z = \pm\frac{3}{2})/\sigma(j_z = \pm\frac{1}{2})$ will have a peak value ~3, which agrees with McGuire. These features are also shown in Fig. 1. Note that for Mg, which does not have a

Cooper minimum, the general behavior of Eq. (21) persists almost to threshold.

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