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Cooper minima: a window on nondipole photoionization at low energy

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Abstract

Photoionization of Mg 3s is studied near the Cooper minimum in dipole channels using the relativistic-random-phase approximation. While the importance of first-order nondipole effects on photoelectron angular distributions at low energies is well known, it is reported here for the first time that in the energy region near the dipole Cooper minimum, quadrupole transitions are not just important, but actually dominate the total photoionization cross section. Studies of dipole–dipole, dipole–quadrupole and quadrupole–quadrupole interference terms in the photoelectron angular distribution show that in the region of dipole Cooper minimum even the calculation of the dipole angular distribution parameter, β , requires the inclusion of quadrupole channels. The significance of second-order [$O(k^2r^2)$] nondipole terms, primarily due to the contributions from electric quadrupole–quadrupole interference terms at photon energy as low as ~ 11 eV, are shown to induce dramatic changes in the photoelectron angular distribution over a small energy range.

It has been generally believed that the absorption by an atom of low-energy photons, having energies up to ~ 5 keV above the ionization threshold, is described rather well by the electric dipole approximation [1–3]. However, at lower energies, the importance of first-order nondipole effects, resulting from interference of electric dipole (E1) and electric quadrupole (E2) ionization amplitudes, has been observed in photoelectron angular distributions [4–8]. The possibility of strong nondipole effects at extremely low photon energy (~ 13 eV) was also noted [9]. With developments in high precision instruments and brighter light sources, however, it has been found that even at much lower energy, hundreds or even just tens of eV, a correct description of the differential photoionization cross section requires the inclusion of first-order, quadrupole, corrections to the dipole approximation [10–16]. In all of these investigations, quadrupole effects have been seen in the photoelectron angular distribution, but have never been found large enough to manifest themselves

in total (integrated) cross sections. This occurs because the quadrupole matrix element, resulting from the expansion of $e^{ik\cdot r}$, includes a factor of k , the photon wave number which is very small at low energy; for a photon energy of 1 Rydberg, for example, $k = \alpha/2$ in atomic units, α being the fine structure constant. Thus, for outer and near-outer subshells, where $r \sim a_0$, the magnitude of the quadrupole photoionization matrix element, which enters the lowest order nondipole correction to the angular distribution linearly, is typically about a factor of 300 smaller than dipole; but for the total cross section, where the correction goes as the absolute square of the quadrupole matrix element, the quadrupole term is about five orders of magnitude smaller than dipole. Of course, near the ionization thresholds of deep inner shells, where the threshold energies are much higher, quadrupole effects are correspondingly relatively larger, of the order of 10% in

angular distributions and 1% in total cross sections [17, 18]⁵. In any case, in the 1 Rydberg photon energy range, quadrupole effects can become important when the quadrupole matrix element is abnormally large, like at a quadrupole resonance [19], or when the dipole matrix element is uncharacteristically small, like at a dipole Cooper minimum.

Recently, significant second-order nondipole effects, primarily due to electric-dipole–octupole and electric-quadrupole–quadrupole interference in the photoelectron angular distributions of 2s and 2p subshells of neon, were demonstrated in the photon energy range of 100–1200 eV [20]. To the best of our knowledge, this is the only case where second-order nondipole effects were ever found to be of importance in this energy range or lower. This prompted us to search for other such situations where second-order terms are important. In this work, significant second-order nondipole effects, due to interference of E2–E2 channels, are demonstrated in the photoionization of the 3s shell of magnesium, at extremely low photon energy (~ 11 eV).

The dipole Cooper minimum, a minimum in the dipole photoionization matrix element [21], was discovered experimentally almost a century ago [22]. Thought originally to be an isolated curiosity, it was found to be a feature of the dipole photoionization matrix element $l \rightarrow l + 1$ transitions of almost all outer and near-outer subshells over the entire periodic table. These minima, located near the ionization threshold in each case, result from the cancellation of the positive and negative contributions to the dipole matrix element. For the photoionization of subshells with $l \neq 0$, the cross section is the sum of the dipole-allowed $l \rightarrow l + 1$ and $l \rightarrow l - 1$ transitions, so that minima are generally not evident in the integrated subshell cross section; even though the $l \rightarrow l + 1$ transition might have a minimum, the $l \rightarrow l - 1$ transition does not. For the photoionization of s-states, simple (independent-particle) theory predicts a zero minimum. This implies that, at a dipole Cooper minimum for an s-state, the cross section arises from quadrupole and higher multipole photoabsorption. This, however, is mitigated by the fact that, even for the photoionization of s-states, the cross section is never zero for two reasons. First, the effect of relativistic interactions causes the $s \rightarrow p_{1/2}$ and the $s \rightarrow p_{3/2}$ minima to occur at slightly different energies, so that the total dipole cross section never vanishes [23, 24]. In addition, correlation in the form of interchannel coupling causes the dipole matrix elements to be complex, and the real and imaginary parts go to zero at slightly different energies, so that the dipole cross section never vanishes, even without relativistic effects [25]. But it is evident that to obtain a quantitatively accurate cross section in the region of the dipole Cooper minimum, or even a qualitatively accurate one, both relativistic and correlation effects must be included in a calculation; a non-relativistic independent-particle calculation which lacks both of these effects simply will not do.

Now, at low photon energies, the quadrupole photoionization cross section is generally far smaller than the dipole cross section, as mentioned. But at a Cooper

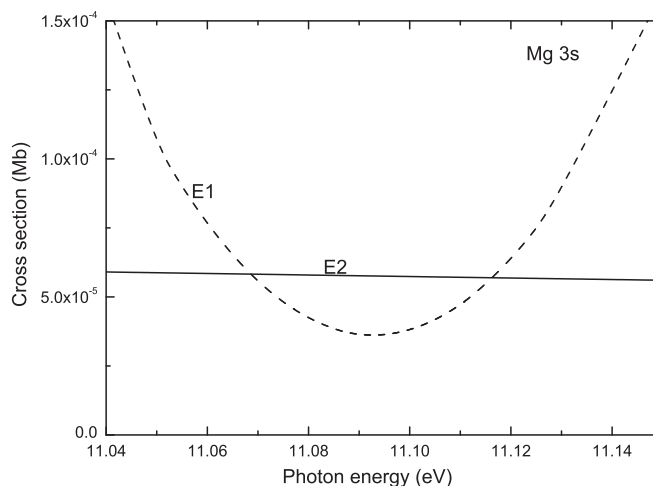


Figure 1. Dipole (E1) and quadrupole (E2) photoionization cross sections for Mg 3s.

minimum in the photoionization of an s-state, it is possible that quadrupole effects could become significant, or even dominant. Note, however, that in all cases that have been investigated, even in the vicinity of a dipole Cooper minimum, the dipole cross section remains far larger than the quadrupole and the integrated subshell cross section is well represented by just the dipole contribution [26, 27]. In this paper, we investigate the possibility that there are some instances where the quadrupole cross section actually does dominate at low energy, a result that would be surprising indeed. To perform this investigation, the relativistic-random-phase approximation (RRPA) [28] has been employed. This methodology includes both relativistic effects and interchannel coupling that are crucial to a qualitatively and quantitatively accurate rendering of the shape and depth of dipole Cooper minima, although the actual energy could be off owing to the use of Dirac–Fock energies in the RRPA along with the neglect of two-electron resonances. We emphasize that, in earlier work using the RRPA, although the threshold energies, as well as the energies of various prominent features of photoionization cross sections, were often significantly off, the dynamics of the cross sections were generally quite accurate [25–29]. In any case, the same formulation has been used to calculate the quadrupole photoionization channels. For both dipole and quadrupole calculations, coupling among single-excitation photoionization channels from all occupied subshells was included.

As an example, the photoionization of the 3s valence subshell of the Mg atom has been studied. To begin with, the results for the dipole and quadrupole total 3s subshell cross sections are shown in figure 1 in the region of the dipole Cooper minimum. This clearly shows that the dipole cross section, although small, does not go through a zero, but still the quadrupole cross section is larger than the dipole over a range of photon energies approximately 0.05 eV. This is the first case found where the quadrupole is larger than the dipole in any realistic calculation; and it is remarkable that it occurs at such a low energy—certainly below where one would ordinarily expect nondipole effects to manifest

⁵ Note that in these references, quadrupole matrix elements are defined, unlike here, without k , which is factored out.

themselves. The inclusion of some other configurations in the initial state may alter the position of the Cooper minimum somewhat. Notwithstanding that possibility, the results of this work clearly and unambiguously demonstrate the importance of the first- and second-order corrections coming from the quadrupole terms in determining the angular distribution of the photoelectrons. In any case, any measurement of the total subshell cross section would only observe the sum of these two cross sections, and this would not provide information on the relative sizes of the dipole and quadrupole cross sections; but observation of the photoelectron angular distribution would. It is important to add that such a measurement is entirely within the capabilities of the present experimental technology.

This work brings out an important aspect regarding the theoretical determination of the dipole angular distribution asymmetry parameter β . This parameter has been studied theoretically in many cases, and in virtually all of these investigations, it has been obtained [30] omitting any nondipole effects. This work, however, demonstrates that nondipole terms need to be included even in the determination of the dipole angular distribution asymmetry parameter, since β is given by [31]

$$\beta = \frac{1}{\bar{\sigma}} \sum_{\kappa\kappa'} \sqrt{30} \langle \kappa' \| C_2 \| \kappa \rangle (-1)^{j'+j_b} \begin{Bmatrix} 1 & 1 & 2 \\ j' & j & j_b \end{Bmatrix} \Re[D_\kappa D_{\kappa'}^*], \quad (1)$$

where $\bar{\sigma} = \sum_{\kappa} [|D_\kappa|^2 + \frac{k^2}{20} |Q_\kappa|^2]$ in the denominator of equation (1) is essentially the total cross section, including the quadrupole part. D and Q in the above denote the dipole and quadrupole matrix elements, respectively, κ' and κ are the final state relativistic quantum numbers corresponding to the transition from the initial state, k is the photon momentum, $\Re[X]$ designates the real part of argument X and $\langle \kappa' \| C_n \| \kappa \rangle$ is the reduced matrix element of the spherical tensor:

$$C_{nq}(\hat{r}) = \sqrt{\frac{4\pi}{2n+1}} Y_{nq}(\hat{r}).$$

In a spectral region where the quadrupole cross section is not negligible, as in the present case, the denominator must include the quadrupole cross section. Thus, in previous cases of the calculation of β where the quadrupole cross section matters, the calculated β must be multiplied by a factor of $\sigma_d/(\sigma_d + \sigma_q)$, where σ_d and σ_q are the dipole and quadrupole subshell cross sections, respectively. Clearly, this factor is always less than (or equal to) unity, so it can only serve to lower the absolute value of β . In the present case, for example, in the vicinity of the Cooper minimum, β drops to a value of about -0.4 ; without the inclusion of the quadrupole part of the cross section, the calculated β would drop to a value of -1.0 (figure 2). Other cases might not be so dramatic, but it would be well to re-examine various results investigated previously where the inclusion quadrupole effects could be of importance in obtaining a quantitatively correct value of β .

The photoelectron angular distribution, the differential cross section, including both dipole and quadrupole

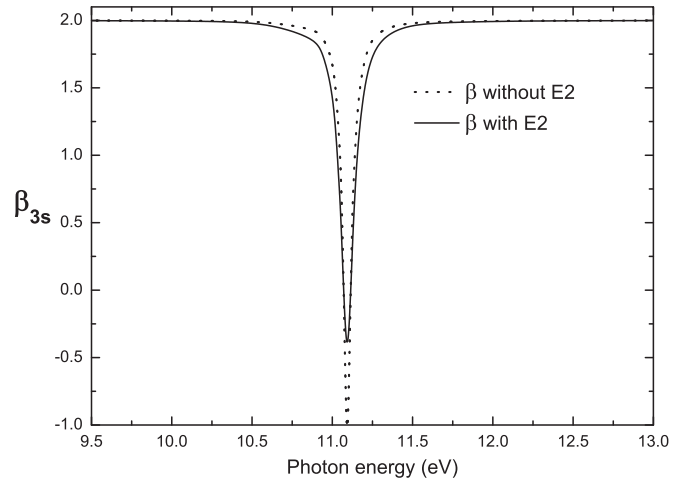


Figure 2. Dipole asymmetry parameter, calculated by considering the effect of E2 (solid line) and without considering E2 (dotted line), in the region of dipole Cooper minimum.

contributions, is given generally for the photoionization of subshell i by linearly polarized photon as [31]

$$d\sigma_i/d\Omega = (\sigma_i/4\pi)[1 + (\beta + \Delta\beta)P_2(\cos\theta) + (\delta + \gamma \cos^2\theta) \sin\theta \cos\varphi + \lambda P_2(\cos\theta) \cos 2\varphi + \mu \cos 2\varphi + \nu(1 + \cos 2\varphi)P_4(\cos\theta)], \quad (2)$$

where σ_i is the total subshell cross section, θ and φ are the polar and azimuthal angles of the photoelectron direction in the coordinate system defined by the photon polarization as the z -axis and the photon direction as the x -axis. In the above equation, $P_2(\cos\theta) = \frac{3\cos^2\theta - 1}{2}$ and $P_4(\cos\theta) = \frac{35\cos^4\theta - 30\cos^2\theta + 3}{8}$ are the second- and fourth-order Legendre polynomials, respectively. The dipole angular distribution parameter β results from dipole–dipole interference, the first-order (in kr) nondipole correction parameters, δ and γ from dipole–quadrupole interference, and the second-order (in kr) nondipole correction parameters, $\Delta\beta$, λ , μ and ν from quadrupole–quadrupole interference; clearly, in the absence of quadrupole effects, the shape of the angular distribution is determined entirely by the β parameter. The detailed expressions for the various parameters are given elsewhere [31] where it is also noted that, for photoionization of an ns-state, the parameter δ is vanishingly small.

The results of our calculations of the angular distribution parameters are shown in figure 3 which exhibits several outstanding features. First is the huge magnitude of the γ parameter in the vicinity of the dipole Cooper minimum. Second are the dramatic changes in γ , as a function of photon energy, in the vicinity of the Cooper minimum. Third is the existence (and rapid change with energy) of the second-order nondipole parameters near the minimum. This is the first indication of non-negligible second-order nondipole contributions to the photoelectron angular distribution below about 1 keV. Furthermore, it is seen that the dipole angular distribution parameter β , which is usually 2 for the photoionization of s-states of a closed-shell atom [30], exhibits a deep dip in the vicinity of the Cooper minimum, reflecting

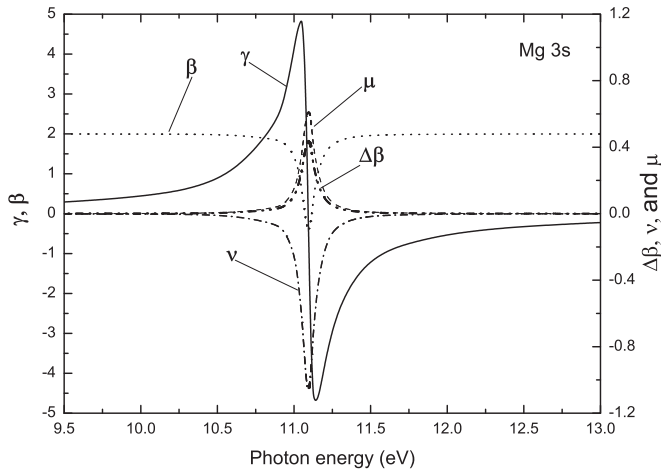


Figure 3. Dipole asymmetry parameter β (dotted line), first-order nondipole asymmetry parameter γ (solid line), and second-order nondipole angular distribution parameters, $\Delta\beta$ (dash-dotted line), μ (dash line) and ν (dash dot line) for Mg 3s in the neighbourhood of the dipole Cooper minimum. The left scale applies to β and γ , and the right scale for the others. The asymmetry parameter $\lambda = -\mu - \nu$ so, for clarity, it is not shown.

the splitting of the minima in the $s \rightarrow p_{1/2}$ and the $s \rightarrow p_{3/2}$ channels. In any case, the strength of the nondipole parameters along with the strong energy dependence of all of the angular distribution parameters means that the angular distribution will also exhibit an extremely strong dependence upon energy.

To demonstrate this dependence, this volatility of the photoelectron angular distribution most simply, the differential cross section, equation (2), is recast as

$$d\sigma_i/d\Omega = (\sigma_i/4\pi)[1 + A(\theta, \varphi)], \quad (3)$$

where $A(\theta, \varphi)$ represents all of the angular information and $1 + A(\theta, \varphi)$ represents the shape of the angular distribution, unencumbered by the overall magnitude of the cross section which also changes rapidly with energy. In figure 4, three-dimensional plots of the shape of the angular distribution, $1 + A(\theta, \varphi)$, for a selection of six photon energies spanning the dipole Cooper minimum are presented and a truly remarkable evolution of the shape is demonstrated. At the lowest photon energy shown, about 9.52 eV, well below the minimum, the angular distribution is seen to be essentially $\cos^2 \theta$ with no dependence on the azimuthal angle φ ; this reflects the situation of $\beta = 2$ and all the nondipole parameters vanishingly small. At about 10.88 eV, still below the minimum, a notable φ dependence is evident, indicating the effects of nondipole interactions, quadrupole in this case. Going up a bit further in energy to approximately 11.02 eV, just below the minimum, the angular distribution changes dramatically and the φ dependence is at least as strong as the θ dependence. This occurs owing to the huge value of the γ parameter just below the minimum, as seen in figure 3. Moving up slightly, to about 11.16 eV, there is another dramatic change in the shape of the angular distribution where the φ dependence dominates and the θ dependence is much less important; this is because the γ parameter goes through a zero at the

Cooper minimum (figure 3) along with the strength of the second-order nondipole parameters. Then, with a further increase in energy, above the Cooper minimum, the angular distribution gradually goes back to the simple $\cos^2 \theta$ shape as the dipole cross section ‘recovers’ from the Cooper minimum and quadrupole effects become proportionally less and less important.

Looking at equation (2), it is evident that *any* φ dependence of the photoelectron angular distribution is a hallmark of nondipole effects, and the very strong φ dependence, along with the very rapid changes of the angular distribution as a function of photon energy, as seen in figure 4, is indicative of very strong nondipole effects. But this is to be expected in a region of the spectrum where the quadrupole photoionization cross section is actually larger than the dipole.

Getting back to the integrated cross sections shown in figure 1, the energies of the two relativistic $s \rightarrow p_j$ dipole Cooper minima shall be designated as C_1 and C_2 for $j = 1/2$ and $3/2$, respectively, separated by the spin-orbit splitting. Since the radial functions for $\epsilon p_{1/2}$ and $\epsilon p_{3/2}$ continua are so similar, the respective dipole matrix elements behave similarly in the vicinity of their respective zeros; in fact, they are linear in a small region which includes both zeros, i.e. $M_{1/2}(E) = \alpha(E - C_1)$ and $M_{3/2}(E) = \alpha(E - C_2)$; the cross sections, proportional to $|M|^2$, are then $\sigma_{1/2} = A\alpha^2(E - C_1)^2$, $\sigma_{3/2} = 2A\alpha^2(E - C_2)^2$, noting that the $s \rightarrow p_{3/2}$ cross section includes an extra factor of 2 compared to $s \rightarrow p_{1/2}$, so that, defining $E_0 = (C_1 + 2C_2)/3$ and $\Delta = (C_1 - C_2)/2$, the total cross section, σ , is given by

$$\sigma = \sigma_{1/2} + \sigma_{3/2} = 3A\alpha^2(E - E_0)^2 + 3A\alpha^2(8/9)\Delta^2, \quad (4)$$

a parabola as a function of photon energy. This can be generalized to include interchannel coupling by taking the dipole matrix elements to be complex where both the real and imaginary parts of the matrix elements go through zeros at slightly different energies; this leads to a more complicated expression, not very different from equation (4) since the imaginary terms are small, and still a parabola, just as is seen in figure 1.

Without loss of generality then, the dipole cross section can be written in a small energy region in the vicinity of the Cooper minima as

$$\sigma_D = F(E_0)[(E - E_0)^2 + (8/9)\Delta^2] \quad (5)$$

for some function $F(E)$. In the small energy region around the dipole Cooper minimum, to an excellent approximation the quadrupole cross section is constant, i.e. $\sigma_Q = K$, for some constant K , assuming that we are not near a quadrupole Cooper minimum (a minimum in the absolute value of the quadrupole photoionization matrix element). Then, in the region of the Cooper minima, the total cross section

$$\begin{aligned} \sigma(E) &= \sigma_D + \sigma_Q = F(E_0)[(E - E_0)^2 + (8/9)\Delta^2] + K \\ &= F(E_0)(E - E_0)^2 + \sigma(E_0) \end{aligned} \quad (6)$$

with $\sigma(E_0) (=F(E_0)(8/9)\Delta^2 + K)$, the cross section at $E = E_0$. Then, if measurements are comprised of E_0 , the bottom of the parabola, along with the cross section at any two energies in the region of the minima, then E_0 , $F(E_0)$ and $\sigma(E_0)$, can be

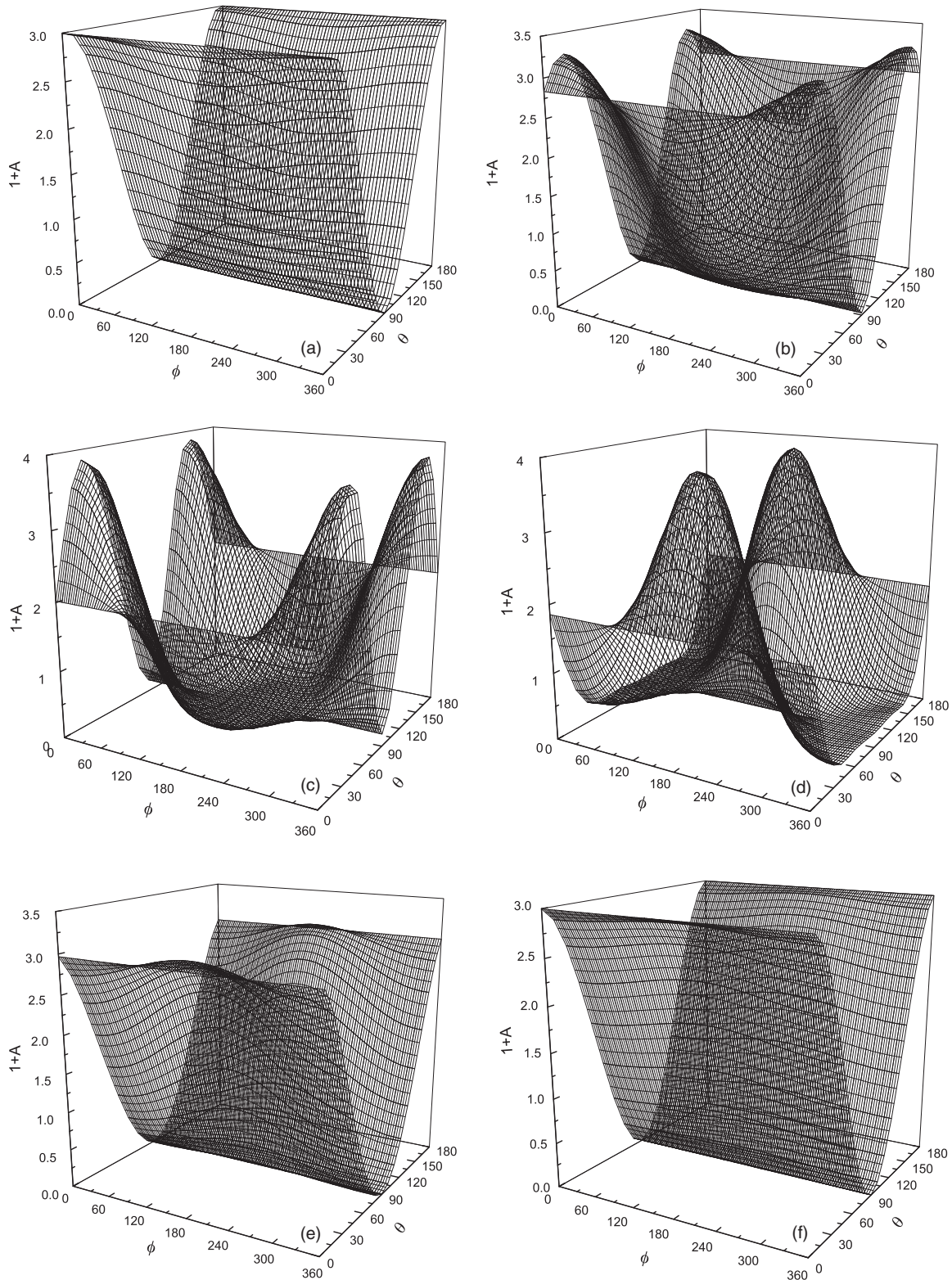


Figure 4. Shape of the photoelectron angular distribution, $1 + A(\theta, \varphi)$, for Mg 3s in the neighbourhood of the dipole Cooper minimum: (a) at photon energy 9.52 eV, (b) photon energy 10.88 eV, (c) photon energy 11.02 eV, (d) photon energy 11.16 eV, (e) photon energy 11.56 eV and (f) photon energy 12.92 eV.

obtained from the measurements. Unfortunately, this does not allow us to obtain separately the quadrupole cross section, K , or the splitting of the dipole Cooper minima, Δ , without further assumptions. However, it does put a useful check on theory

owing to the relationship among E_0 , $F(E_0)$, Δ , K and $\sigma(E_0)$, i.e. along with the three measurements, if theory does a good job of predicting the positions of the dipole Cooper minima (and thereby Δ), then the quadrupole cross section, K , could

be obtained without any quadrupole calculation. On the other hand, if K in the region could be accurately calculated, then Δ could be obtained and, thus, the positions of the relativistic dipole Cooper minima. In any case, this analysis shows that scrutiny of the photoionization cross section in the vicinity of the dipole Cooper minima leads to a relationship that provides a stringent test of theory which is not obtainable in any other manner.

In summary then, Mg 3s photoionization is shown to exhibit very strong effects of quadrupole photoionization in the vicinity of the Cooper minimum, effects which show up particularly dramatically in the shape and evolution of the photoelectron angular distribution. Thus, even in a photon energy region of the order of 10 eV, large and rapidly changing nondipole parameters, both of first and second orders, are found. And this should also be the case for the photoionization of other atoms with a 3s Cooper minimum, from Na to Ar, and to a lesser extent, owing to stronger relativistic interactions, for heavier atoms with a 4s Cooper minimum, starting with K. Using a simple relativistic independent-particle calculation, we have confirmed that the same effects occur for other elements in the Na to Ar row of the periodic table. It is also likely that similar effects will occur in regions of the dipole resonance series, since between the resonances the dipole cross section can drop to a very small value [32]. This should be the most important at low Z where relativistic interactions are small. And, at somewhat higher energies, inner shells, even deep inner shells, might offer opportunities to examine quadrupole effects in the vicinities of the small dipole cross sections between the resonances of a Rydberg series, especially because, as noted earlier, quadrupole cross sections are ordinarily larger, compared to that of the dipole, for inner shells. These findings suggest a new arena for laboratory investigations, studies of photoelectron angular distributions in the vicinity of dipole minima, which are within the capabilities on modern experimental technology with regard to energy resolution [33] and sensitivity to small cross sections [34]. Regardless of the cross section of the order of 10^{-5} Mb being measurable or not, it is clear that the effect on angular distribution is measurable. The dramatic changes shown in figure 3 are attributed to the fact that $\sigma_{E2} > \sigma_{E1}$ in the vicinity of the dipole Cooper minimum.

In addition, calculations of the dipole angular distribution parameter β , particularly for the ns-state photoionization in the vicinity of Cooper minima, or other energy regions where dipole cross section might be quite small, need to be re-examined for possible effects of quadrupole photoionization. Finally, it is shown that under most conditions, the shape of the cross section in the vicinity of the dipole Cooper minima is parabolic and the addition of the quadrupole cross section, which is essentially constant over a small energy region around the dipole Cooper minimum, does not change that fact; it would be well to investigate photoionization cross section in the region of Cooper minima experimentally which would provide the most stringent test of theory.

Acknowledgments

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