Krypton E2 photoionization cross-section and angular distribution of photoelectrons

Taniman Banerjee\(^1\), P C Deshmukh\(^1\) and S T Manson\(^2\)

\(^1\)Department of Physics, Indian Institute of Physics - Madras, Cheenai 600036, India
\(^2\)Department of Physics and Astronomy, Georgia State University, Atlanta, GA-30303, USA

E-mail: tanima@physics.iitm.ac.in

Abstract. A systematic study of electric quadrupole (E2) photoionization electronic transitions from \(4p\), \(4s\) and \(3d\) subshells for atomic krypton is carried out in the photon energy range from their respective ionization thresholds up to photon energy of 1500 eV. Truncated relativistic random phase approximation (RRPA) is employed at different levels of interchannel coupling to study the effect of relativistic interactions and electron correlations. It is found that the E2 angular distribution asymmetry parameters are rather sensitive to interchannel coupling.

1. Introduction

In the regime of the ‘dipole approximation’ [1] the valence shell photoionization for atomic krypton has been studied in details both experimentally and theoretically at different levels of approximation [2-6]. In recent years with developments in high precision spectroscopy, it is now possible to measure even small contributions from higher order terms in \(\frac{e}{\hbar}\) which lead to ‘non-dipole’ photoionization transitions [7-16]. Inclusion of the first order term in \(\frac{e}{\hbar}\) leads to electric quadrupole (E2) transitions and magnetic dipole (M1) transitions. However, the magnetic dipole transition amplitudes are very weak and can be neglected. The higher order cross-sections being proportional to absolute square of transition matrix elements are very small and hence are difficult to measure. The angular distribution asymmetry parameters however depend on the matrix elements themselves and are amenable to reliable measurements [9].

The RRPA [17] formalism to determine the E2 photoionization cross-section and angular distribution parameters was developed by Johnson et al. [10, 11]. There are hitherto no detailed experimental or theoretical studies on the E2 transitions for atomic ‘krypton’. In the present work we have extended the earlier [10] theoretical studies on non-dipole photoionization of valence shell for atomic krypton using truncated RRPA which allows selective inclusion of electron correlations by coupling channels from \(4p\), \(4s\), \(3d\), \(3p\), \(3s\), \(2p\), \(2s\) at different levels. Computations have been carried out in photon energy range from the respective ionization thresholds up to photon energy \(\sim 1500\) eV. The focus of the present study is on the study of the dynamics of ‘Cooper minimum’ [18] in E2 channels.

2. Results and Discussion

The differential cross-section for photoionization from the \(i^{th}\) subshell is given by [19]

\[
\frac{d\sigma_i}{d\Omega} = \frac{\sigma_i}{4\pi} [1 + \beta_i P_2(\cos \theta) + (\delta_i + \gamma_i \cos^2 \theta) \sin \theta \cos \phi],
\]
where $\sigma_i$ is the angle-integrated cross-section, $\beta_i$ is the dipole angular distribution asymmetry parameter which arises from E1-E1 interference terms, and $\delta_i$ and $\gamma_i$ are non-dipole asymmetry parameters which arise from E1-E2 interference terms; $\theta$ and $\phi$ are polar coordinates of the photoelectron momentum vector $\vec{p}$ in a coordinate system with photon polarization vector along z-axis and photon propagation vector along $x$-axis.

In the present work we have used the experimental ionization thresholds from Ref.[10], which enables the inclusion of some non-RPA correlations [20], for different subshells of atomic krypton listed in Table 1.

**Table 1.** Experimental thresholds for atomic krypton

| Subshells | Thresholds in eV |
|-----------|------------------|
| $4p_{\frac{3}{2}}^3$ | 14.0002 |
| $4p_{\frac{1}{2}}^3$ | 14.6996 |
| $4s$ | 27.4998 |
| $3d_{\frac{5}{2}}$ | 94.2004 |
| $3d_{\frac{3}{2}}$ | 95.4004 |
| $3p_{\frac{3}{2}}$ | 214.7985 |
| $3p_{\frac{1}{2}}$ | 222.3986 |
| $3s$ | 293.0992 |
| $2p_{\frac{1}{2}}$ | 1679.1002 |
| $2p_{\frac{3}{2}}$ | 1730.8998 |
| $2s$ | 1920.3998 |
| $1s$ | 14327.1008 |

Electric quadrupole photoionization parameters were studied using RRPA at different levels of truncation, coupling channels from the outer most subshell $4p$ down to $2s$ in the photon energy range from the respective ionization thresholds up to 1500 eV. Except channels from the $1s$, interchannel coupling with all the subshells were included, thus providing a 'minimal' truncation of the RRPA. In the following sections E2 photoionization parameters the cross-section ($\sigma$) and the angular distribution asymmetry parameters ($\gamma$ and $\delta$) are reported for $4p$, $4s$ and $3d$ subshells of atomic krypton.

2.1. E2 photoionization cross-section

RRPA studies have been carried out at the following two levels of truncation:

(i) coupling channels from intrashell.

(ii) coupling all the channels from $4p$, $4s$, $3d$, $3p$, $3s$, $2p$, $2s$ subshells.

In Figure 1 is plotted the E2 cross-section for 4p subshells at the two levels of truncation of RRPA mentioned above. At intrashell coupling level (i), the $4p_{\frac{3}{2}}^3$ and $4p_{\frac{1}{2}}^3$ E2 cross-sections go through Cooper minima at $\sim 95$ eV [Figure 1(a)]. The relativistic effects are very weak, not surprisingly since krypton is not a very heavy atom and cross-sections for both the $4p_{\frac{3}{2}}^3$ and $4p_{\frac{1}{2}}^3$ subshells go through Cooper minimum at almost the same photon energy. Furthermore, at the intrashell coupling level, at higher photon energy, the cross-sections go through another Cooper minimum $\sim 900$ eV [Figure 1(b)]. For the second Cooper minimum the relativistic effects are not as weak as for the first Cooper minimum. The three E2 channels $4p_{\frac{3}{2}}^3 \rightarrow \epsilon f_{\frac{1}{2}}^3$, $4p_{\frac{3}{2}}^3 \rightarrow \epsilon f_{\frac{3}{2}}^3$ and $4p_{\frac{1}{2}}^3 \rightarrow \epsilon f_{\frac{1}{2}}^3$ go through the second Cooper minimum at photon energies 881 eV, 877 eV and
Figure 1. Kr 4p E2 cross-section at two levels of truncation of RRPA (i) coupling 6 channels from 4p intrashell coupling and (ii) coupling 33 channels from 4s, 4p, 3d, 3p, 3s, 2p, 2s in the photon energy range (a) From 4p ionization threshold up to 200 eV; (b) from 200 eV to 1500 eV. Vertical lines show the ionization threshold energies for the various subshells of atomic Kr.

918 eV respectively. The Cooper minimum in $4p_{\frac{3}{2}} \rightarrow \epsilon f_{\frac{5}{2}}$ channel is at a lower photon energy than $4p_{\frac{1}{2}} \rightarrow \epsilon f_{\frac{3}{2}}$ channel, since the spin-orbit interaction for $j = l - \frac{1}{2}$ is attractive whereas it is repulsive for $j = l + \frac{1}{2}$ [21]. Therefore, the continuum $\epsilon f_{\frac{3}{2}}$ wavefunction is more compact than $\epsilon f_{\frac{5}{2}}$ wavefunction. This makes the $4p_{\frac{3}{2}} \rightarrow \epsilon f_{\frac{5}{2}}$ channel go through the Cooper minimum at lower photon energy than the minimum in $4p_{\frac{1}{2}} \rightarrow \epsilon f_{\frac{3}{2}}$ channel. Furthermore, due to similar reasons, Cooper minimum in channel $4p_{\frac{3}{2}} \rightarrow \epsilon f_{\frac{5}{2}}$ occurs at a higher energy than the minimum in $4p_{\frac{1}{2}} \rightarrow \epsilon f_{\frac{3}{2}}$ channel. One might expect that the splitting between the Cooper minimum in the two channels with the same final state is equal to the $4p_{\frac{3}{2}} - 4p_{\frac{1}{2}}$ spin-orbit splitting of bound state energies. However, the splitting in the position of the Cooper minima is rather large, ~41 eV, whereas the spin-orbit splitting of the bound state energies for 4p is ~0.7 eV. This is due to the fact that the centrifugal barrier for f waves make it more difficult for the continuum f waves to penetrate to the core region than for the discrete p orbitals. The strength of the centrifugal barrier for f waves is responsible for this huge separation in the position of the Cooper minima. Multiple Cooper minima have been reported in earlier studies on photoionization of excited states of atomic cesium [22] and also in some cases of photoionization from the ground state of high Z atoms where multiple minima are induced by interchannel coupling [23, 24]. In the present study, since the second minimum is seen even in the truncated RRPA computations performed at level (i), it is clear that multiple Cooper minima can be found even in the case of photoionization from ground state without interchannel coupling.

The position of the first Cooper minimum is not sensitive to interchannel coupling. However, this is not the case for the second Cooper minimum at the higher photon energy (~900 eV). When additional channels are coupled (33 channels), the second Cooper minima occur in $4p_{\frac{3}{2}} \rightarrow \epsilon f_{\frac{5}{2}}$, $4p_{\frac{1}{2}} \rightarrow \epsilon f_{\frac{3}{2}}$ and $4p_{\frac{1}{2}} \rightarrow \epsilon f_{\frac{1}{2}}$ channels at 895 eV, 890 eV and 930 eV respectively. Comparison of the positions of the Cooper minima in the channels at the two levels of truncation of RRPA clearly indicates a shift of the Cooper minimum in each channel to a slightly higher photon energy due to interchannel coupling.

Just above the 4s ionization threshold there are some small but significant effects of interchannel coupling on 4p E2 cross-section that have been noticed, but not shown in Figure 1(a). This behavior of the 4p cross-section above the 4s threshold will be studied in details and reported later.

Figure 2(a) shows the E2 cross-section of the 4s subshell at the two levels of truncation
(a) Cross-section in 3d channels coupling 33 channels from 4s, 4p, 3d, 3p, 3s, 2p, 2s in the photon energy range from 3d ionization threshold up to 800 eV. (b) Kr 3d E2 cross-section in truncated RRPA coupling 33 channels from 4p, 4s, 3d, 3p, 3s, 2p, 2s from 3d ionization threshold up to 800 eV. Vertical lines show the ionization threshold energies for the various subshells of atomic Kr.

mentioned above. At the intrashell coupling level the cross-section goes through a Cooper minimum at \( \sim 66 \text{ eV} \). When additional channels are coupled the position of the Cooper minimum shifts to higher photon energy, \( \sim 78 \text{ eV} \). The 4p cross-section is about 10 times stronger than the 4s cross-section near the Cooper minimum in 4s. Therefore, interchannel coupling with channels from 4p subshells shifts the position of E2 Cooper minimum in 4s channels significantly. An analysis of the partial cross-sections in the individual E2 channels \( 4s \to ed_{z^2} \) indicates that the relativistic effects on the position of the Cooper minimum is very weak. Both the channels and also the total cross-section of 4s subshell go through the minimum at almost same photon energy at each level of truncation. Also, the value of the cross-section in 4s subshell at the Cooper minimum is almost equal to zero which again is due to the fact that both the 4s channels go through their respective Cooper minimum at very nearly the same photon energy.

In Figure 3(a) is shown the E2 cross-section of 3d subshell using RRPA with level (ii) of

truncation. The interchannel coupling effects are not significant in this case. The E2 cross-
sections shown in Figure 3(a) go through a ‘dip’ near the ionization threshold. In Figure 3(b) is shown the cross-sections in channels $3d_{\frac{3}{2}} \rightarrow eg_{\frac{1}{2}}$ and $3d_{\frac{5}{2}} \rightarrow cd g_{\frac{5}{2}}$. The $3d \rightarrow cd$ matrix elements decrease monotonically above the threshold. However, $3d \rightarrow eg$ matrix elements rise from the ionization thresholds and subsequently go through a ‘shape resonance’. Due to this difference in energy dependence of the channels, near the ionization threshold $3d \rightarrow eg$ channels dominate the cross-section profile and with increase in photon energy it is the $3d \rightarrow cd$ channels which become dominant. The ‘dip’ in the E2 cross-section is due to this difference in the energy dependence of $3d \rightarrow eg$ and $3d \rightarrow d$ relativistic channels. It may be added that the E1 cross-section of the $3d$ subshell was also found to show a similar ‘dip’ near the ionization thresholds due to the difference in energy dependence of $3d \rightarrow ef$ and $3d \rightarrow ep$ relativistic channels [25]. The actual energy profile of the cross-section depends on the relative strengths of the individual channels.

2.2. E2 angular distribution asymmetry parameter

E2 angular distribution asymmetry parameters $\gamma$ and $\delta$ are obtained from the interference of E1 and E2 transition amplitudes. The dynamics of $\gamma$ and $\delta$ is therefore influenced by both E1 and E2 transition amplitudes. The expressions [10, 11] for $\gamma$ and $\delta$ in RRPA methodology are rather complicated.

In Figure 4 are shown the E2 angular distribution asymmetry parameters $\gamma$ and $\delta$ for $4p$ subshells at the two levels of truncation of RRPA. As mentioned above, the expressions for $\gamma$ and $\delta$ [10, 11] are rather complicated and hence making any straightforward interpretation of
their energy dependence is difficult. For s-subshell photoionization, however, the non-relativistic expression for γ is amenable for a relatively straightforward interpretation. In non-relativistic single particle approximation for ns photoionization, γ and δ take a rather simple form [19]:

\[
\gamma = 3\alpha \omega \frac{Q_{\omega,d}}{D_{\omega,p}} \cos(\delta_{\omega,p} - \delta_{\omega,d}),
\]

\[
\delta = 0,
\]

where \(\alpha\) is the fine structure constant, \(\omega\) is the photon energy, \(D_{\omega,p}\) and \(Q_{\omega,d}\) are respectively the electric dipole and quadrupole radial matrix elements, \(\delta_{\omega,p}\) and \(\delta_{\omega,d}\) are the final state phase shifts for E1 and E2 transitions. Figure 5 shows the angular distribution asymmetry parameter γ of 4s subshell. In Figure 5 at the level of intrashell coupling, γ is maximum at the ionization threshold. When E1 channels go through a Cooper minimum, the ratio in Equation 2 of E2 to E1 matrix elements becomes large. It has been found that at the intrashell level of coupling of RRPA, the Cooper minimum in 4s E1 channels occurs below the ionization threshold (which agrees with the result in [26]). The large value of γ (intrashell) at the threshold is therefore due to this E1 Cooper minimum. The exact value of γ depends on the ratio of the E2 and E1 matrix elements and also on the cosine term. Near \(\sim 66\) eV there is a Cooper minimum in E2 channels \((Q_{\omega,d} \approx 0)\) which manifests as \(\gamma \approx 0\). Also, at \(\sim 29\) eV of photon energy \(\gamma \approx 0\), whereas at this photon energy \(Q_{\omega,d} \neq 0\). This indicates that the zero in γ near photon energy of \(\sim 29\) eV is due to the cosine term.

When additional channels are coupled at the level (ii) coupling of RRPA, the γ profile gets modified dramatically. It has been found from the E1 RRPA results that due to coupling of additional channels from 4p subshell, the E1 Cooper minimum in 4s shifts to an energy above the ionization threshold (which is in agreement with the earlier studies reported in [26]). γ therefore increases from zero at the ionization threshold and goes through a peak \(\sim 42\) eV of photon energy which is the region of E1 Cooper minimum. Subsequently, near the Cooper minimum in E2 channels \(\sim 78\) eV (Figure 2), γ goes through a zero. Also, there is a zero in γ near \(\sim 50\) eV where the quadrupole channels does not go through any Cooper minimum. Therefore, the cosine term would be zero, making γ ≈ 0. Thus, at a Cooper minimum in E2 channel, γ goes through a near zero, however it is not necessary that if γ is zero the E2 channels would also go through a minimum.

In Figure 6 is shown the the angular distribution asymmetry parameters γ and δ at the level (ii) coupling of RRPA. It has been found that the energy profile of γ and δ at both the levels of truncation of RRPA mentioned above is almost identical and hence in Figure 6, results of truncation (i) are not shown. As in the case of 4p photoionization, the energy profiles of γ and δ are very complicated.

Figure 5. Kr 4s E2 angular distribution asymmetry parameter γ at two levels of truncation of RRPA (i) coupling channels from 4s subshell; intrashell coupling and (ii) coupling channels from 4s, 4p, 3d, 3p, 3s, 2p, 2s subshells. Vertical lines show the ionization threshold energies for the various subshells of atomic Kr.
3. Conclusions
The dynamics of E2 matrix elements near the Cooper minimum is sensitive to relativistic and correlation effects, even if not as dramatic as in the E1 case. The E2 photoionization channels for 4p subshells go through two Cooper minima even at the level of intrashell coupling. Although, the first Cooper minimum is not sensitive to interchannel coupling, the second minimum shifts to relatively higher photon energy due to interchannel coupling. In the case of 4s E2 photoionization there is a single Cooper minimum and it shifts to relatively higher energy due to interchannel coupling with channels from 4p subshells. In photoionization of 3d subshell there is no Cooper minimum since 3d wavefunction is nodeless. However, the 3d cross-sections go through a ‘dip’ near the ionization thresholds due to the difference in the energy dependence of the individual channels. The energy profiles of the E2 angular distributions for 4p and 3d subshells are too complex, however the energy dependence of the 4s angular distribution is relatively simple. It is hoped that the present results would stimulate some experimental measurements of the non-dipole photoionization parameters of atomic krypton.

Acknowledgments
This work was partially supported by BRNS (DAE), DST (India) and NSF (USA). The authors are grateful to Professor Walter Johnson (University of Notre Dame) for valuable discussions and for providing us his RRPA codes.

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