Interchannel coupling effects in the spin polarization of energetic photoelectrons

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Effects of the interchannel coupling on the spin polarization of energetic photoelectrons emitted from atomic Ne valence subshells are examined. Like previously obtained results for cross sections and angular distributions, the photoelectron spin polarization parameters too are found considerably influenced by the coupling. The result completes a series of studies to finally conclude that the independent particle description is inadequate for the entire range of photoionization dynamics over the full spectral energy domain.

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I. INTRODUCTION

The study of the spin polarization of photoelectrons is important primarily on two counts. Firstly, since this effect originates from purely relativistic interactions, its behavior provides insights into relativistic aspects of the dynamical correlation which are inaccessible by conventional studies of cross section and angular distribution; in the photoionization of lighter atoms, which may normally seem tractable by a non-relativistic approach, sizable resonance features in spin polarization spectra of emerging electrons can be found when relativistic forces are included.[1, 2] Secondly, spin resolved spectroscopic measurements in conjunction with cross section and angular distribution data provide a comprehensive methodology to completely characterize the photoionization process.[3, 4] As a virtual beginning of the interest in the field, the emission of highly spin polarized electrons over a limited range of the ejection angle near the Cooper minimum of the photoionization cross section was predicted by Fano many years ago.[5] Since then, the spin polarization of photoelectrons emanating from unpolarized atoms has been the subject of several theoretical and experimental investigations (for reviews see Refs. [6, 7]). However, the focus of all these studies has been the low photon energy range (VUV and soft X-ray), over which it is a common knowledge that the electron correlation, in its complete form including interchannel coupling, is significant and often dominating.

On the other hand, at photon energies far away from the ionization threshold, it was believed until recently that the independent particle (IP) framework, which completely disregards electron correlation, can adequately describe the photoionization process.[8, 9, 10, 11, 12, 13] However, over a series of combined experimental and theoretical studies this notion has recently been corrected for the cross section and the angular distribution asymmetry parameter.[14, 15, 16] Interchannel coupling has been shown to be a crucial determinant of the quantitative accuracy of these parameters in the intermediate and the high energy regime for photoelectrons emitted from both inner and outer atomic subshells. From a perturbative perspective, the effect originates from the correction to the single channel matrix element from a continuum configuration interaction among all neighboring channels.[17]. Since the dynamical difference among relativistic (spin-orbit) channels arising from a given subshell determines the spin polarization character of electrons photoejected from that subshell, it is of particular interest to examine how the spin polarization parameters are affected by correlation in the form of interchannel coupling mechanism. In this paper, we focus on this aspect by investigating the spin polarization parameters of the valence 2p photoelectrons from atomic Ne.

II. ESSENTIAL THEORETICAL DETAILS

The relativistic-random phase approximation (RRPA)[17, 18] has been employed to perform the calculation. The RRPA calculation starts from an explicitly relativistic basis so that relativistic interactions are included ab initio. In addition to the ground state correlation, as well as two-electron promotion in the residual Ne-ion core, RRPA incorporates interchannel coupling among all of the single excitation/ionization final state channels. We use a framework in which the coupling among selective members of the relativistic dipole-allowed $jj$-coupled channels:

$$1s_\frac{1}{2} \rightarrow kp_\frac{1}{2}, kp_\frac{3}{2},$$
$$2p_\frac{1}{2} \rightarrow kd_\frac{1}{2}, ks_\frac{1}{2},$$
$$2s_\frac{3}{2} \rightarrow kp_\frac{3}{2}, kp_\frac{5}{2},$$
$$2p_\frac{3}{2} \rightarrow kd_\frac{3}{2}, kd_\frac{5}{2}, ks_\frac{3}{2}$$

(1)

can be chosen. The calculation has been carried out in both the length and the velocity gauge formalism; the good agreement between length and velocity results, even at highest photon energy considered, indicates the numerical accuracy of our calculation. It may be mentioned...
here that RRPA has been used previously with reasonable success to study the spin polarization of photoelectrons from noble gases, but only in the low photon energy range\textsuperscript{19}.

In this calculation, we have considered the case where the target atom is unpolarized and the polarization of the residual ion is not observed. Equivalently, the polarization of the target atom is averaged out and that of the residual ion is summed over. The dipole photoionization can then be completely described, in general, by a set of five dynamical parameters $\sigma$, $\beta$, $\xi$, $\eta$, and $\zeta$, wherein $\sigma$ is the partial cross section, $\beta$ is the angular distribution asymmetry parameter and the others are the photoelectron spin polarization parameters. These dynamical parameters can be expressed in terms of the reduced dipole matrix elements\textsuperscript{17} of the process. The explicit expressions for the spin polarization parameters corresponding to both $2p_{1/2}$ and the $2p_{3/2}$ photoelectrons of Ne are given as $20$

\[
\xi_{2p_{1/2}} = \left[ \frac{1}{2} |D_{S1/2}|^2 + \frac{2}{5} |D_{d_{1/2}}|^2 - \frac{9}{10} |D_{d_{5/2}}|^2 - \frac{\sqrt{5}}{2} |D_{S1/2}||D_{d_{3/2}}| \cos\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) \right] \times \sigma_{2p_{1/2}}^{-1}
\]

(2a)

\[
\xi_{2p_{3/2}} = \left[ - |D_{S1/2}|^2 + |D_{d_{5/2}}|^2 - \frac{1}{\sqrt{2}} |D_{S1/2}||D_{d_{3/2}}| \cos\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) \right] \times \sigma_{2p_{3/2}}^{-1}
\]

(2b)

\[
\eta_{2p_{1/2}} = \left[ \frac{3}{2} \sqrt{\frac{1}{5}} |D_{S1/2}||D_{d_{3/2}}| \sin\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) + 3 \sqrt{\frac{1}{5}} |D_{S1/2}||D_{d_{5/2}}| \sin\left(\theta_{S1/2} - \theta_{d_{5/2}}\right) \right] \times \sigma_{2p_{1/2}}^{-1}
\]

(3a)

\[
\eta_{2p_{3/2}} = \left[ \frac{3}{\sqrt{2}} |D_{S1/2}||D_{d_{3/2}}| \sin\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) \right] \times \sigma_{2p_{3/2}}^{-1}
\]

(3b)

\[
\zeta_{2p_{1/2}} = \left[ - \frac{1}{2} |D_{S1/2}|^2 + \frac{1}{5} |D_{d_{3/2}}|^2 + \frac{3}{10} |D_{d_{5/2}}|^2 - \frac{\sqrt{5}}{2} |D_{S1/2}||D_{d_{3/2}}| \cos\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) \right] \times \sigma_{2p_{1/2}}^{-1}
\]

(4a)

\[
\zeta_{2p_{3/2}} = \left[ |D_{S1/2}|^2 + \frac{1}{2} |D_{d_{3/2}}|^2 - \frac{\sqrt{2}}{2} |D_{S1/2}||D_{d_{3/2}}| \cos\left(\theta_{S1/2} - \theta_{d_{3/2}}\right) \right] \times \sigma_{2p_{3/2}}^{-1}
\]

(4b)

where, with the photon energy $\omega$, the subshell cross sections are

\[
\sigma_{2p_{1/2}} = \frac{8\pi^4}{\omega c} \sigma_{2p_{1/2}} = \frac{8\pi^4}{\omega c} \left[ |D_{S1/2}|^2 + |D_{d_{3/2}}|^2 + |D_{d_{5/2}}|^2 \right]
\]

(5a)

\[
\sigma_{2p_{3/2}} = \frac{8\pi^4}{\omega c} \sigma_{2p_{3/2}} = \frac{8\pi^4}{\omega c} \left[ |D_{S1/2}|^2 + |D_{d_{3/2}}|^2 \right]
\]

(5b)

In the above equations we use the short-hand notations $D_{ij}$ and $\theta_{ij}$, for the reduced matrix element $D_{ni_j \rightarrow kl_{j'}}$, and the phase-shift $\theta_{ni_j \rightarrow kl_{j'}}$, respectively, corresponding to the $nl_j \rightarrow kl_{j'}$ dissociation channel. Conventionally,
additional parameters $\delta_{nlj} = (\zeta_{nlj} - 2\zeta_{nlj})/3$ are also used which connect to the spin polarization of the total photoelectron flux. 20

III. RESULTS AND DISCUSSION

In order to uncover the details of how the interchannel coupling influences the photoelectron spin polarization dynamics we have performed five separate calculations for the 2p$_{\pm 2}$ and 2p$_{\pm 3}$ photoionization of Ne with varying degrees of interchannel coupling. These are (i) no interchannel coupling among channels arising from different relativistic subshells, (ii) coupling of all channels from 2p$_{\pm 2}$ and 2p$_{\pm 3}$ subshells, (iii) from 2p and 2s subshells, (iv) from 2p and 1s subshells, and (v) from all four subshells together (full calculation). Since, in calculation (i), we ignore all effects of interchannel coupling between channels arising from differing relativistic subshells, this is similar to the relativistic independent particle (IP) description, except that ground state correlations are included along with coupling among channels arising from the same subshell.

An elegant approach to understand the relative importance of the coupling with different neighboring channels has been described in Ref. [14] in the spirit of a first order perturbation theory. Under the influence of a perturbing degenerate channel $J$ the corrected wavefunctions $\Psi_{2p_j}$ for any dipole channel $2p_j \rightarrow ks_{1/2}(kd_{j'})$ (see Eq. (1)) from either of $2p_j (j = 1/2, 3/2)$ subshells are given, at the photoelectron kinetic energy $E$, by

$$\Psi_{2p_j}(E) = \psi_{2p_j}(E) + \int \frac{\langle \psi_J(E') | H - H_0 | \psi_{2p_j}(E) \rangle}{E - E'} \psi_J(E')dE',$$

(6)

where $\psi$’s denote unperturbed wavefunctions, which are eigenfunctions of the unperturbed Hamiltonian $H_0$, and the final state total angular momentum $j'$ has two dipole allowed values 5/2 and 3/2. In Eq. (6), the matrix element under the energy integration is the interchannel coupling matrix element with $H$ being the full Hamiltonian of the system. Now, defining the dipole photoionization matrix element for $2p_j \rightarrow ks_{1/2}(kd_{j'})$ transitions with no interchannel coupling among channels arising from different relativistic subshells, corresponding to calculation (i), as

$$D_{2p_j}(E) = \langle \psi_I | T | \psi_{2p_j}(E) \rangle,$$

(7)

with $\psi_i$ being the ground state wavefunction and $T$ the transition operator, the corresponding perturbed matrix elements can be expressed as

$$M_{2p_j}(E) = D_{2p_j}(E) + \int \frac{\langle \psi_J(E') | H - H_0 | \psi_{2p_j}(E) \rangle}{E - E'} D_J(E')dE'.$$

(8)

The correction term on the right side of Eq. (5) can be significant if two conditions are simultaneously satisfied. First, the spatial overlap between the perturbed and perturbing channel wavefunctions must be considerable to result in a significant interchannel coupling matrix element; this is expected when the discrete wavefunctions have the same principle quantum numbers so that they occupy the same region of space and have significant overlap, and the respective ionization thresholds are close so that at high enough energies the electrons from both subshells have similar momenta, which enable the continuum wavefunctions to oscillate roughly “in phase”. Second, the magnitude of the unperturbed matrix element of the perturbing channel is considerably larger than that of the perturbed channel. Now the form of the energy integral suggests that the primary contribution of interchannel interaction will come from the values of the integrand at $E' \approx E$. Importantly further, the electron continuum wavefunctions, participating in the energy integral, must be normalized per unit energy through a multiplication by a factor $mh^{-2} E^{-1/2}$ [21]. Therefore, the leading energy behavior of the interchannel coupling matrix element in Eq. (6) turns out to be $E^{-1/2}$ when the energy is high enough. Evidently, considering Eq. (8), if the uncoupled matrix element $D_J$ of channel $J$ decreases with energy slower by a factor $E^{1/2}$ or more than the corresponding decay of $D_{2p_j \rightarrow ks_{1/2}(kd_{j'})}$, the resulting effect of the coupling will be considerable, provided the interchannel coupling matrix element is significant. Indeed, this leads us to expect a strong effect of the 2s channels (with Dirac-Fock threshold 52.68 eV) on $2p_j$ photoionization (with thresholds 23.08 eV for $j = 3/2$ and 23.21 eV for $j = 1/2$). This is clearly seen in Fig. 1 which gives our calculated results for the $2p_{1/2}$ and the $2p_{3/2}$ subshell cross sections in each of the five calculations described above; at the highest energies, the cross section results are seen to essentially coalesce into just two curves - those including coupling between and those omitting that coupling.
The weak effect of 1s channels on either spin-orbit subshell cross sections, as also seen in Fig. 1, is owing to the much higher 1s ionization threshold (893.02 eV) that results in poor overlap of the continuum wavefunctions in ensuing interchannel coupling matrix element, along with the fact that the discrete 1s and 2p\textsubscript{j} occupy very different regions of space and, therefore, overlap poorly.

In addition, what is rather interesting to note in Fig. 1 is the tiny effect from the coupling between the channels from the spin-orbit split 2p\textsubscript{j} subshells. The curve (thin solid) corresponding to only-2p\textsubscript{3/2} or only-2p\textsubscript{1/2} channels (calculation (1), similar to the IP result) differs very little in the lower part of the energy range (Fig. 1(a)) when compared to the curve (dotted) from all 2p channels combined; however, both the curves practically merge together at higher energies (Fig. 1(b)) indicating virtually no effect from the coupling. This phenomenon can be understood as follows. It is true that the interchannel coupling matrix element between the 2p\textsubscript{3/2} and 2p\textsubscript{1/2} is strong due to the close proximity of their respective ionization thresholds. But since the high energy falloff of \( D_{2p_{3/2}} \) and \( D_{2p_{1/2}} \) are the same, the ensuing coupling corrections (Eq. (8)) fall off effectively as \( E^{-1/2} \) at higher energy, explaining how the small coupling effect at lower energies becomes practically zero at higher energies.

Looking at our results for the spin polarization parameters, Figs. 2 through 5, rather different phenomenology is evident; significant effects resulting from the coupling between the channels arising from the 2p\textsubscript{3/2} and 2p\textsubscript{1/2} spin-orbit subshells are noted! Understanding the underlying reason(s) for this phenomenology is somewhat more complex than for the cross sections owing to the fact that the spin polarization parameters depend upon both the magnitudes and the phases of the dipole matrix elements, as seen from Eqs. (2-4). Thus, an understanding of the modification of the phase shifts engendered by interchannel coupling is also of importance.

Starting from Eq. (8) and explicitly introducing the unperturbed \( \langle \Theta \rangle \) and the perturbed \( \langle \Theta \rangle \) phase-shift through the notations \( D = |D| \exp(i \langle \Theta \rangle) \) and \( M = |M| \exp(i \langle \Theta \rangle) \) we obtain

\[
\Theta_{2p_j} = \theta_{2p_j} - i \log \left[ \left( D_{2p_j} \right) (E) + \int \frac{\langle \psi_j(E') \vert H - H_0 \vert \psi_{2p_j}(E) \rangle}{E - E'} |D_j| \exp[i(\theta_j - \theta_{2p_j})](E')dE' \right] \times |M_{2p_j}|^{-1}. \tag{9}
\]

Thus, as discussed above, sufficiently above the ionization thresholds the leading energy behavior of \( \langle \psi_j \vert H - H_0 \vert \psi_{2p_j} \rangle \) is \( E^{-1/2} \). From Eq. (9) this indicates the decreasing effect of the coupling on the relative phase-shift going up in the energy. However, it is important to note here that this decay is much slower than \( E^{-1/2} \) due to the logarithmic nature of the correction — a behavior which bears some consequence in the interchannel coupling effects on the photoelectron spin polarization parameters.

Let us first focus on the effect of 2p\textsubscript{3/2} channels on the spin polarization of 2p\textsubscript{1/2} photoelectrons and \textit{vice versa} (dotted curves). We compare between the thin solid curve (effectively the IP prediction) and the dotted curve.
of each of the Figs. 2 through 5. Evidently, for all the spin polarization parameters the effect of this coupling is stronger at relatively low energies. This is clearly because the strength of the interchannel coupling matrix element decreases with increasing energy, as discussed above. But, as already made clear from the corresponding cross section results, this coupling does not strongly affect the magnitude of the $2p_j$ dipole matrix elements. And since their unperturbed phases are nearly equal, the coupling does not alter their phases much (see Eq. (9)). As a consequence, our results (not shown) of the angular distribution asymmetry parameter $\beta$, that depends on both the matrix elements and the phases, shows minimal effect of this coupling. But if the coupling affects neither the dipole matrix elements nor the phases significantly, how can it affect the spin polarization parameters? The answer lies in the fact that the values of the $2p_j$ spin polarization parameters arise from complicated combinations of the dipole matrix elements and their phases, Eqs. (2-4), resulting in significant cancellations, so that small differences in dipole matrix elements and phases can be magnified to produce the results seen. The fact that the non-relativistic limit of spin polarization parameters are zero while for the $\beta$ parameter it is finite and close to its relativistic value indicates that such a cancellation mechanism is indeed operative for the spin polarization parameters.

Interchannel coupling of the $2p$ channels with either $2s$ or $1s$ channels involves alterations in both the magnitudes and phases of the $2p$ dipole matrix elements. Coupling with the $2s$ channels affects the magnitudes of the $2p_{3/2}$ and $2p_{1/2}$ dipole matrix elements very strongly, as clearly indicated in Fig. 1. The phases are also strongly affected (see Eq. (9)). With the $1s$ coupling on the other
hand, while the modifications to the magnitudes of the 2p\textsubscript{j} dipole matrix elements are already small, except in a very small region around the 1s threshold, the alterations of the 2p\textsubscript{j} phase shifts are also small. But it is non-trivial to assess in which direction the changes in these dynamical quantities, the magnitudes and the phases of the 2p\textsubscript{j} dipole matrix elements, will induce changes in the spin polarization parameters. For two of the spin polarization parameters, ξ (Fig. 2) and δ (Fig. 5), the effect of 2s and 1s coupling on 2p\textsubscript{j} ionization is certainly not as straightforward as in the case of cross sections where only the magnitudes of the matrix elements (and not the phases) are important. In addition, by virtue of the different functional dependence of the parameters on the magnitudes of the dipole matrix elements and phase-shifts, the qualitative behavior of the result changes from one parameter to another.

As seen in Fig. 2, the effect of 1s and 2s coupling on 2p\textsubscript{j} for the parameter ξ are roughly complementary until about 900 eV photon energy, the position of 1s Rydberg resonances; as a result, the deviation between the dotted (all 2p channels included) and thick solid (full calculation) results remain approximately constant. Beyond the resonance region the relative effect of 1s coupling drops off. However, over the entire energy range considered, the full calculation differs from the effective IP result (thin solid curve). This difference is significant for 2p\textsubscript{3/2} showing a maximum alteration of about 25% at 300 eV, while for 2p\textsubscript{1/2} the difference is rather small.

For parameters η and ζ, on the other hand, the 1s and 2s coupling influence the result quite in the similar qualitative manner as they do for the cross section. Along almost the complete energy range, albeit the near-threshold region (where interchannel coupling is known to be important), η for both 2p\textsubscript{3/2} and 2p\textsubscript{1/2} (Figs. 3(a,b)) as well as 2p\textsubscript{3/2} ζ (Fig. 4(a)) suggest an almost steady coupling contribution of more than 25% over the corresponding effective IP prediction. For 2p\textsubscript{1/2} ζ (Fig. 4(b)), of course, the effect is small at lower energies which, however, increases gradually with energy to yield over 20% correction.

The parameter δ (Fig. 5) being a combination of parameters ξ and ζ exhibits a rather mixed behavior. For 2p\textsubscript{3/2} photoionization (Fig. 5(a)) a maximum of 40% coupling effect is seen at around 300 eV that monotonically diminishes with increasing energy to eventually produce about 20% effect over the high energy range. For 2p\textsubscript{1/2} (Fig. 5(b)) the effect of the coupling, that is weak at low energies, rises steadily to reach a value of about 20% at the highest energy considered.

These results clearly demonstrate that, as in the case of cross sections and angular distributions, the effect of interchannel coupling on the photoelectron spin polarization is considerable. However, due to the sensitivity of the spin polarization to relative phase-shifts the results exhibit behavior that is qualitatively different from that of the cross section results. While for the cross section,
the high energy interchannel coupling effect generically increases going from lower to higher photon energies, for most of the spin polarization parameters a strong coupling contribution appears already at low energies. As a consequence, a substantial coupling correction exists for these parameters over a very broad spectral range. Furthermore, coupling with 1s channels, which influences the cross section only over a narrow range, extends its influence over a much larger energy range for the 2p, spin polarization parameters owing to the much slower drop-off of the phase shift corrections induced by interchannel coupling. Finally, we note that although we have illustrated the effect with an example of valence photoionization of Ne, the same mechanism of configuration interactions in the continuum (interchannel coupling) must be operative for the spin-polarization of photoejected electrons from any arbitrary atom or atomic ion and from any subshell.

IV. CONCLUSION

It is shown in this paper that the photoelectron spin polarization of an atom is strongly influenced by the electron correlation via interchannel coupling over the entire spectral range. Unlike to the cross section, where only the coupling-induced alteration of the magnitudes of matrix elements is responsible for the behavior, modification of the phase-shifts play an important role determining the effect on the spin polarization. For the cross section and the angular distribution the importance of interchannel coupling for energetic photoemission has been demonstrated previously [14]. With the current result we conclude, therefore, that in order to acquire a complete knowledge of photoionization dynamics unambiguously over the range from VUV all the way to hard x-rays, theoretical study including the interchannel coupling is absolutely required.

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