Relativistic effects on interchannel coupling in atomic photoionization: The photoelectron angular distribution of Xe 5*s*

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Measurements of the photoelectron angular-distribution asymmetry parameter β for Xe 5s photoionization have been performed in the 80–200 eV photon-energy region. The results show a substantial deviation from the nonrelativistic value of $\beta=2$ and provide a clear signature of significant relativistic effects in interchannel coupling.

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Although the importance of interchannel coupling in the photoionization of atoms and ions was recognized some time ago [1–3], a recent upsurge of activity in this area has occurred [4–12]. This has served to reemphasize and extend the region of importance of interchannel coupling on the photoionization process. More specifically, while the earlier work focused on effects in the threshold region, including inner-shell thresholds [1–3], this later work has shown that the importance of interchannel coupling is far more general than that. Recent examples show the significance of interchannel coupling far above threshold and away from inner-shell thresholds [4–6,10], at high energy where the asymptotic form of the nonrelativistic cross section is altered [9], and in quadrupole channels [11,12], and, in some cases, the interchannel coupling dominates the transition matrix element [7]. The essential element in all of these cases is that the interchannel coupling (configuration interaction in the continuum) modifies the transition matrix elements of weak channels that are degenerate with strong ones. As a result of all of this recent activity, it has been found that interchannel coupling is of importance for *most subshells of most atoms at most energies* [4–12].

An excellent way to highlight interchannel-coupling effects is to study photoelectron angular distributions because interferences among strong and weak channels are often determinative. This is because the variation of the angular distribution with energy is a result of the variation of the interferences among the alternative final channels leading to a particular final state of the photoion. This is reflected in the energy dependence of the angular-distribution asymmetry parameter β_i, which, for 100% linearly polarized incident radiation, is related to the differential cross section in the dipole approximation by [13–15]

$$\frac{d\sigma_i}{d\omega}=\frac{\sigma_i}{4\pi}[1+\beta_i P_2(\cos\theta)], \quad (1)$$

where i is the designation of the final state of the photoion, σ_i is the integrated cross section, θ is the angle between photoelectron-momentum and photon-polarization directions, and $P_2(x)=(3x^2-1)/2$ is the Legendre polynomial of order 2.

For photoionization of ns electrons, in the simple central-field approximation, β is constant and equal to 2 because only a single $s\rightarrow p$ transition is possible; with only a single continuum wave, no interference can occur. In an open-shell atom this can be modified owing to the possibility of various couplings of the ϵp continuum wave with the open-shell ionic core, multiple partial waves are possible, and these partial waves interfere with each other [13]. In certain cases, the difference among the various partial waves can be due to interchannel coupling as seen recently in the case of Sc 4s photoionization [8]. In closed-shell systems, the situation is different; only relativistic effects can provide a breakdown of the $\beta=2$ behavior by allowing the possibility of differences between the $s\rightarrow p_{1/2}$ and $s\rightarrow p_{3/2}$ transition amplitudes. This is known to occur near threshold due to exchange effects, near Cooper minima for which the two relativistic transition amplitudes go through zero at different energies, in the vicinity of resonances that occur selectively in one relativistic channel or the other, or at very high energies where j-dependent relativistic effects become large. In this paper it is pointed out that it can also occur due to relativistic effects in interchannel coupling well above threshold, which can cause the $s\rightarrow p_{1/2}$ and $s\rightarrow p_{3/2}$ transition amplitudes to differ.

Up to this point, there have been a number of theoretical studies of relativistic effects causing a deviation of an ns β from 2, almost all in the noble gases [12,16–19]. However, only one case, Xe 5s in the near-threshold region, has been studied in detail experimentally [16,17]. In this case, the deviation was found to result from a combination of exchange effects and interchannel coupling of the 5s channels with 4d, 5p, and 5p satellite channels [20–22].

In fact, calculations that omit interchannel coupling entirely still show a significant deviation from $\beta=2$ in the 5s threshold region. The introduction of interchannel coupling changes the energy at which this deviation occurs [12,22]. It is thus difficult to pinpoint the effects of interchannel cou-

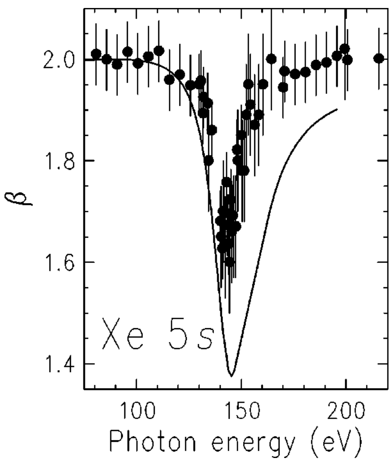

FIG. 1. Photoelectron angular-distribution asymmetry parameter β for the $5s$ subshell of Xe well above threshold. The points are the present experimental results, and the solid curve is the theoretical result [12].

pling alone. In this paper, an experimental measurement of β is presented, also in Xe $5s$, but far above threshold where exchange effects are completely unimportant [8], which unequivocally shows the effects of relativistic interactions in interchannel coupling.

The experiments were performed at the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory on undulator beam line 8.0 using a gas-phase time-of-flight (TOF) photoelectron-spectroscopy system designed specifically for soft-x-ray work at the ALS. A complete discussion of this apparatus is published elsewhere [23]. A key characteristic for the present measurements is that the TOF method can measure photoelectron peaks at many kinetic energies and at multiple emission angles simultaneously, permitting sensitive determinations of cross-section ratios and electron angular distributions with minimal experimental uncertainty.

It was important to separate the Xe $5s$, $5p$, and satellite lines for accurate measurements of the $5s$ angular distributions. Therefore, a retarding voltage between 0 V and -90 V was applied to slow the electrons at higher photon energies. The neon $2s$ photoline was used to calibrate the analyzers which were located in the plane perpendicular to the photon propagation direction at $\theta=0°$ and $\theta=54.7°$, the magic angle, with θ defined as in Eq. (1). It was also used to determine the degree of linear polarization of the synchrotron light to better than 99.9%. In addition, the experimental geometry is such that the angular-distribution measurements are performed in the plane where the lowest-order corrections to the dipole angular distribution vanish [23].

The experimental results are shown in Fig. 1, where a significant deviation from $\beta=2$ is evident. As discussed above, for a closed-shell system, this deviation is a signature that the dynamics (radial matrix elements) of $5s\rightarrow\epsilon p_{1/2}$ and $5s\rightarrow\epsilon p_{3/2}$ transitions differ significantly. The photoelectron angular-distribution asymmetry parameter β is given for this case, in the dipole approximation, as [13,24]

$$\beta=\frac{2R_{3/2}{}^2+4R_{3/2}R_{1/2}\cos(\delta_{3/2}-\delta_{1/2})}{2R_{3/2}{}^2+R_{1/2}{}^2} \qquad (2)$$

where $R_{3/2}$ and $R_{1/2}$ are the radial matrix elements for the $5s\rightarrow\epsilon p_{3/2}$ and $5s\rightarrow\epsilon p_{1/2}$ photoionizing transitions, respectively, and $\delta_{3/2}$ and $\delta_{1/2}$ their respective phases. It is evident from this expression that, if $R_{3/2}=R_{1/2}$ and $\delta_{3/2}=\delta_{1/2}$, $\beta=2$ and does not vary with energy.

The substantial variation of β with photon energy seen in Fig. 1 shows a broad shallow minimum. This minimum in β is attributed to interchannel coupling, and differential interchannel coupling at that; the $5s\rightarrow\epsilon p_{1/2}$ and $5s\rightarrow\epsilon p_{3/2}$ transitions are affected differently. It is a general rule that interchannel coupling can dramatically alter the transition matrix elements of a weak channel that is degenerate with a strong one [4–12]. Thus, since the dominant channels in this energy range arise from $4d$ photoionization, the interchannel coupling of the weak $5s$ channels with the strong $4d$ channels is most likely responsible for the broad minimum seen in β_{5s}.

To confirm this interpretation, the results of a relativistic random phase approximation (RRPA) calculation [12] are also shown in Fig. 1. Interchannel coupling is included and the broad minimum is accounted for reasonably well. The calculation also shows that the interchannel coupling of $5s$ with $4d$ channels results in an ''induced'' Cooper minimum [1,3,25] in σ_{5s} in this energy region as well. That this broad minimum is due to interchannel coupling with $4d$ channels is confirmed since omitting this coupling from the calculation results in the broad minimum vanishing and β_{5s} being very close to 2 and virtually constant [12]. This case, therefore, differs from the near-threshold structure in β_{5s}, discussed above, in that, without the introduction of interchannel coupling, β_{5s} is absolutely flat and equal to 2 in the higher-energy region, with no hint of a minimum [12]. Note also that the $4d$ photoionization channels exhibit their Cooper minima around 175 eV [16,17], well above the ''induced'' Cooper minimum in the $5s$ channel.

The agreement between RRPA theory and the present experiment for β_{5s} is much better than the agreement between the two in the threshold region [22], mentioned previously, where the omission of interchannel coupling with $5p$ satellite channels was decisive. In the present case, agreement on the low-energy side is quite good, but on the high-energy side the theoretical result is significantly deeper and broader than experiment. This must also be due to the omission of interchannel coupling with satellite channels, possibly $4p$ satellites in this case, which, although they do not have as dominant an effect as the $5p$ and $5p$ satellite channels had in the threshold region, might still have an effect, as seen in Fig. 1. Alternatively, despite the decrease in the importance of interchannel coupling with the $5p$ satellite channels in going from the $5s$ threshold to the 140 eV region because the interchannel coupling matrix elements fall off as $1/E$ asymptotically [9], it might still be that omission of the $5p$ satellite channels is responsible for the small discrepancy. Or perhaps even the $4d$ satellite channels are important here. On this point we cannot be sure.

In conclusion, measurements of the photoelectron angular-distribution parameter β for the $5s$ subshell of Xe well above threshold have spotlighted relativistic effects on interchannel coupling and the existence of an induced Cooper minimum in the cross section. Reasonable agreement was found with a recent RRPA calculation [12]. Interchannel coupling with $5p$ satellite channels, which have a dominant effect on β_{5s} near the $5s$ threshold [22], are much less important far above the $5s$ threshold. Most importantly, this study demonstrates the information that can be gleaned concerning relativistic effects in a closed-shell atom, through investigation of the β parameters for ns subshells, and is by no means limited to Xe $5s$.

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