Outstanding Spin-Orbit-Activated Interchannel Coupling in the Cs and Ba 3d Photoemission

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(Received 30 December 2006; published 3 April 2007)

Partial $3d_{5/2}$ photoionization cross sections of atomic Cs and Ba as well as the asymmetry parameter β of the angular distribution of the Cs $3d_{5/2}$ photoelectrons were investigated near the threshold of the $3d_{3/2}$ channel at about 750 eV and 800 eV, respectively. Strong electron correlations, in particular, the spin-orbit activated interchannel coupling between the $3d_{5/2}$ and $3d_{3/2}$ channels, govern the observed spectra. The most striking effect was found for $\beta_{5/2}$ of Cs with a dramatic increase from $\beta = 1.0$ to $\beta = 1.5$ in the energy region where the mixing between both channels causes a pronounced minimum in the partial $3d_{5/2}$ cross section. This result indicates the decisive influence of the interference term on the asymmetry parameter β with its dependence on the phase difference between the outgoing p and f waves.

DOI: 10.1103/PhysRevLett.98.143002 PACS numbers: 32.80.Fb

Atomic photoionization is an ideal testing ground to study many-body phenomena which manifest themselves in strong electron correlations and coupling effects. The correct quantum mechanical description of the complex mutual interactions between the individual electrons and between the electrons and the nucleus is the main challenge for all theoretical approaches. The importance of interchannel coupling on the atomic photoionization for outer and inner electrons over a wide range of kinetic energies is generally accepted (see, for example, [1,2] and references therein). Extensive investigations have been performed for resonant excitations, for example, in the case of atomic Xe showing the strong coupling of the outer $5s$ and $5p$ photoionization cross sections to the strong $4d \rightarrow \epsilon f$ shape resonance [3–5]. In addition, angular distribution and spin polarization of photoelectrons are very sensitive parameters to highlight interchannel coupling effects causing, for example, large energy shifts of the maxima of the asymmetry parameter β with respect to the resonance positions, e.g., Diehl *et al.* [6]. In general, pronounced effects occur when the transition matrix elements of a weak channel are modified by mixing with a strong channel. They can be observed in the photoionization spectra of all states of matter from atoms to solids.

More recently, a completely new type of interchannel coupling has been discussed for the $3d$ photoionization of Xe, Cs, and Ba [7] where in the vicinity of the $3d_{3/2}$ threshold the $3d_{5/2}$ channel is strongly influenced by mixing with the $3d_{3/2}$ channel. It was demonstrated that the underlying mechanism is of explicit many-body nature and is activated by the spin-orbit interaction breaking the degeneracy of the $3d$ doublet. The theoretical investigations of these interchannel coupling effects in the $3d$ spin-orbit doublets of Xe, Cs, and Ba by Amusia *et al.* [7–9] using the framework of spin-polarized random phase approximation with exchange (SPRPAE) were stimulated by a detailed experimental study on the $3d$ photoionization of Xe by Kivimäki *et al.* [10] who found an unexpected additional maximum in the $3d_{5/2}$ partial cross section in the vicinity of the $3d_{3/2}$ threshold. Amusia *et al.* [7] explained this feature by interchannel coupling between the continuum states of the $3d$ doublets. The authors [7–9] alleged that the effect of spin-orbit activated interchannel coupling should show up in other atomic and molecular systems and even in clusters and condensed matter. They presented extensive calculations of the partial cross sections, asymmetry parameters, and spin polarization parameters of the $3d$ photoionization of Xe, Cs, and Ba. Their predictions on the partial cross section $\sigma_{5/2}$ of Cs were experimentally confirmed by Farrokhpur *et al.* [11] who observed a pronounced minimum above threshold followed by a second maximum near the $3d_{3/2}$ threshold very similar to the case of Xe.

The main focus of the present study lies in the determination of the asymmetry parameter β in the $3d_{5/2}$ photoionization of Cs in the vicinity of the $3d_{3/2}$ threshold. A pronounced variation of the β value is observed proving in this way the theoretical analysis, which revealed a dramatic increase from 1.0 to 1.5 for $\beta_{5/2}$ in this energy region. This is in contrast to the predictions on the asymmetry parameter for Xe, where the many-body calculations yielded extremely weak variations and only small deviations from the single particle Hartree-Fock (HF) results.

In addition, this Letter reports on measurements of the partial photoionization cross section $\sigma_{5/2}$ of Cs and Ba. Our experimental findings verify qualitatively the theoretical predictions [7–9], which suggest an interpretation of the results for Ba completely different from the above discussion on Xe and Cs. This is due to the collapse of the Ba $4f$ wave function into the inner region which causes a tremendous increase in the overlap with the innershell $3d$ wave function ([12] and references therein). As a result, the $3d$ resonance has moved below threshold into the discrete region. Therefore, the Ba $3d_{5/2}$ partial cross section below the $3d_{3/2}$ threshold is dominated by interchannel coupling with the huge $3d_{3/2} \rightarrow 4f$ autoionizing resonance. In the discussion, we will concentrate on the different influence of the spin-orbit activated interchannel coupling on the partial cross section and the angular distribution of the photoelectrons.

The differential cross section for photoionization of unpolarized (unoriented) atoms with linearly polarized radiation and within the dipole approximation is given by

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{\sigma}{4\pi}[1 + \beta P_2(\cos\theta)].$$

Here θ is the angle between the polarization vector of the ionizing light and the momentum vector of the ejected photoelectron, P_2 is the second Legendre polynomial, σ is the isotropic angle-integrated cross section, and β is the asymmetry parameter describing the angular distribution of the photoelectrons.

A Scienta SES-2002 electron analyzer was used to separate energetically the different photoelectrons. For the determination of the partial cross section only electrons emitted under the magic angle ($54° \, 44'$) with respect to the linear polarization vector of the synchrotron radiation were observed. The intensity of the $3d_{5/2}$ photoline was recorded as a function of the exciting photon beam with proper normalization to the photon flux and the energy dependent transmission of the analyzer. For the measurement of the asymmetry parameter $\beta_{5/2}$ of Cs we used the ability of the BESSY UE52/SGM beam line to rotate the polarization axis of the linearly polarized undulator radiation. In this way electrons emitted perpendicular to the propagation direction of the synchrotron direction were detected at four different angles relative to the polarization axis. Possible quadrupole effects in the angular distribution are thus eliminated. In addition to the measurements of the photoelectrons, the ion yield of the differently charged photoions was detected by a time of flight (TOF) spectrometer. The angle-integrated ion signal can also be used as a reliable standard for the normalization of the angle-resolved photoelectron spectra. A detailed description of the experimental setup and the advantages of a fixed-in-space electron analyzer combined with the variable polarization of the synchrotron radiation have been published elsewhere [13]. The energy scale for the Cs spectra was established measuring well-known Xe Auger and photo lines. For the Ba data the maximum of the $3d_{5/2}$ cross section was set to 799.2 eV, which is the value given for the maximum of the Ba $3d_{5/2}$ absorption ([Xe] $3d^{-1}(^2D_{3/2})6s^2 4f \; ^1P_1$ line) in [12].

For a first overview of the $3d$ excitation region in Cs we have measured partial ion yields of differently charged photoions Cs^{n+} from $n = 1$ to $n = 7$ in the region of 730 eV to 770 eV photon energy (Fig. 1, upper panel). Ions with $n = 4$ and 3 are found to be the major contributors, created in an initial formation of a $3d$ hole and a

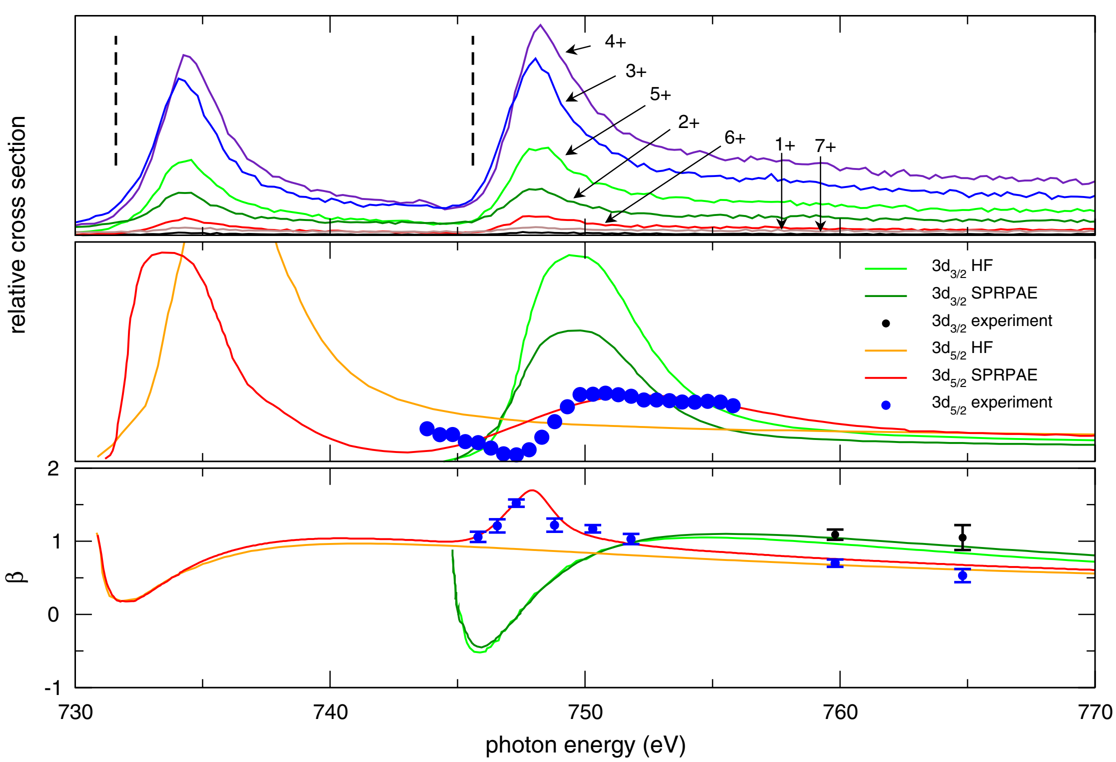

FIG. 1 (color online). (upper panel) Partial ion yield spectra of differently charged photoions Cs^{n+} from $n = 1$ to 7. The two dashed lines indicate the $3d_{5/2}$ and $3d_{3/2}$ thresholds (731.6 eV and 745.6 eV, from [14]). (middle panel) Partial Cs $3d_{5/2}$ photoelectron cross section, together with theoretical values for $\sigma_{5/2}$ and $\sigma_{3/2}$ calculated by HF and SPRPAE, taken from [7]. (lower panel) Cs $3d_{5/2}$ and $3d_{3/2}$ photoelectron asymmetry parameter, together with theoretical HF and SPRPAE values from [8].

number of subsequent Auger decays. The characteristic shape of the ion yield curves with pronounced maxima just above the $3d_{3/2}$ and $3d_{5/2}$ ionization thresholds reflects the influence of the centrifugal barrier "seen" by the f electron waves, weakening the photoionization cross section at threshold. Similar to the situation in Xe, the maxima of the $3d$ photoionization are observed about 3 eV above the thresholds.

The partial cross section $\sigma_{5/2}$ and the asymmetry parameter $\beta_{5/2}$ of Cs are depicted in Fig. 1 (middle and lower panel) together with calculated values of σ and β for both channels $3d_{3/2}$ and $3d_{5/2}$ [7,8]. The energy position of the theoretical cross sections had to be shifted by 4.7 eV, as the calculation was based on solid state threshold data. Qualitative agreement is found with the SPRPAE calculations, showing the strong increase of the $3d_{5/2}$ cross section just above the $3d_{3/2}$ threshold. However, the detailed comparison of the resonances reveals some differences. The shape of the experimental curve shows an almost steplike behavior, whereas theory predicts a broad resonance profile with the maximum slightly above the resonance positions of the ion yield curves.

Our data for the asymmetry parameter $\beta_{5/2}$ confirm the theoretically predicted strong increase of the β value. Also the two data points recorded for $\beta_{3/2}$ agree nicely with the SPRPAE calculation and support the consistency of the results. But in order to match this quite sharp feature in $\beta_{5/2}$, the energy calibration of the calculated β data were shifted by only 4.0 eV, not 4.7 eV as for σ. So the maximum of the $\beta_{5/2}$ values is at lower photon energies than theoretically expected. The different shifts applied to the theoretical σ and β have been chosen for presentation purposes in order to enable a better comparison to the experiment, but they indicate also that further theoretical improvements are necessary to completely describe the observed dependences.

The resonance character of $\sigma_{5/2}$ and $\beta_{5/2}$ is compared in more detail in Fig. 2. It is quite obvious that they behave in a very different way. Whereas the experimental maximum of $\sigma_{5/2}$ is reached at about 749 eV which coincides with the ion yield peak of the $3d_{3/2}$ channel (Fig. 1), the maximum of the $\beta_{5/2}$ values reaches its maximum at or near 747 eV which coincides with the minimum of $\sigma_{5/2}$. While this is true for the experimental data, in the calculated curves the extrema do not match. Note that the energy scale difference 0.7 eV in the theoretical data, discussed above, would not change the context for the SPRPAE data: with the broad partial cross section maximum the β feature would be located in the middle of the σ slope in any case.

For the reason of the distinct behavior of $\sigma_{5/2}$ and $\beta_{5/2}$, one has to reconsider the general remark in the introduction that pronounced interchannel effects appear when the transition matrix elements of a weak channel is modified by mixing with a strong channel. The origin of the difference between $\sigma_{5/2}$ and $\beta_{5/2}$ is the different dependence of both

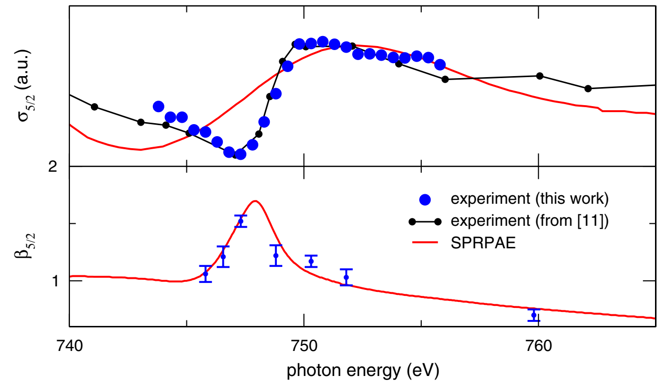

FIG. 2 (color online). (upper panel) Partial Cs $3d_{5/2}$ photoelectron cross section, together with the SPRPAE calculation [7] and experimental data from [11]. (lower panel) Cs $3d_{5/2}$ β parameter measurement and the SPRPAE values [8].

quantities on the transition matrix elements d_p and d_f which describe the transitions $nd \rightarrow \epsilon p$ and $nd \rightarrow \epsilon f$ of the $3d$ electrons into the outgoing p and f waves of the photoelectrons. Whereas the partial cross section σ depends only on the (absolute) square of the transition matrix elements, the asymmetry parameter β as a ratio of the transition matrix elements depends also on the phases of the outgoing electron waves. Although the interchannel coupling effects on β are of explicit many-body nature, the key features can already be deduced from the well-known Cooper-Zare formula. This formula can be used to calculate β for the photoionization of d electrons in the one-electron dipole approximation by

$$\beta = \frac{2d_p^2 + 12d_f^2 - 36d_p d_f \cos(\delta_f - \delta_p)}{5(2d_p^2 + 3d_f^2)}$$

with δ_p and δ_f the scattering phases of the outgoing p and f waves. The crucial point for our discussion is the interference term in the numerator with the cosine dependence of the phase difference between the p and f waves. If the phase difference $|\delta_f - \delta_p|$ is about 90° or one of the two transitions $nd \rightarrow \epsilon p$ or $nd \rightarrow \epsilon f$ dominates, the interference term can be neglected. In that case β behaves as the ratio of the (absolute) squares of the transition matrix elements and the β values approximate $\beta = 0.2$ (ϵp channel dominates) or $\beta = 0.8$ (ϵf channel dominates). In both cases the influence of mixing is very small. If, on the other hand, both transition matrix elements are of the same order of magnitude, the interference term plays the decisive role for the β values. For phase differences $\delta_f - \delta_p$ near 0° or 180° the interference term has its strongest influence on the β values which then approximate $\beta = 2$ for $\delta_f - \delta_p = 180°$ or $\beta = -0.88$ for $\delta_f - \delta_p = 0°$.

Now we consider the effect of interchannel coupling on $\sigma_{5/2}$ and $\beta_{5/2}$. It consists essentially of the mixing of the continuum wave functions of the $3d_{5/2}$ channel with those of the $3d_{3/2}$ channel thereby changing the transition matrix elements and the scattering phases δ_p and δ_f. For the

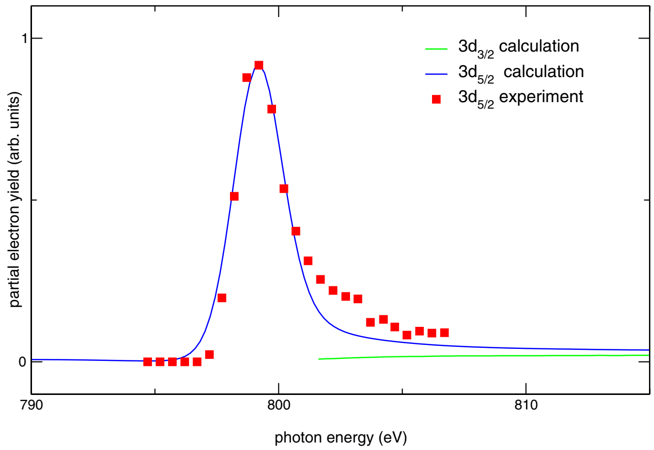

FIG. 3 (color online). Partial Ba $3d_{5/2}$ photoelectron cross section and SPRPAE calculations for $\sigma_{5/2}$ and $\sigma_{3/2}$ taken from [7].

partial cross section $\sigma_{5/2}$ these changes have a pronounced effect in the vicinity of the $3d_{3/2} \rightarrow \epsilon f$ shape resonance with its maximum at 749 eV resulting in the (second) maximum at this photon energy. As the photoionization of the $3d_{5/2}$ channel is dominated by transitions to the ϵf channel in this region the influence of interchannel coupling on $\beta_{5/2}$ is therefore very small. At the minimum of $\sigma_{5/2}$ at 747 eV, on the other hand, there is a sharp increase of $\beta_{5/2}$ indicating that there the transition matrix elements d_p and d_f are of the same order of magnitude and the phase difference $\delta_f - \delta_p$ should be near 180°.

Our measured partial cross section $\sigma_{5/2}$ is in excellent agreement with the recent experimental results of Farrokhpour *et al.* [11], also depicted in Fig. 2. Note the perfect match of the differently established energy scales. The minor deviations in the trend at the ends could be attributed to slight normalization problems in either of the data sets. This independent verification of the steplike $\sigma_{5/2}$ profile calls for an improved calculation.

Furthermore, we have measured the $3d_{5/2}$ photoionization cross section of Ba in the region around the $3d_{3/2}$ threshold at about 800 eV excitation energy (Fig. 3). The solid lines represent the SPRPAE calculations by Amusia *et al.* [7], which have been convoluted by a Gaussian distribution of 1.9 eV width to match the experimental resolution and have been shifted to overlap in the cross section maximum at 799.2 eV. Experimental and theoretical curves are in reasonable agreement demonstrating thereby the strong interchannel coupling effect also for the $3d_{5/2}$ cross section in Ba. But this observed resonance is of completely different origin than for Xe and Cs, since the resonance maximum is situated below threshold. The shape of the resonance profile is almost symmetric and closely resembles the form of the absorption curve [12]. The collapse of the $4f$ wave function upon $3d$ excitation causes the strong discrete $3d_{3/2} \rightarrow 4f$resonance, which is seen in the $3d_{5/2}$ cross section. Unfortunately, the proposed strong changes in the β value [8] coincides with a vanishing $3d_{5/2}$ cross section. That made an experimental verification of the theoretical β results impossible and leaves it as an extremely challenging task for the future.

In conclusion, our measurements of partial cross sections $\sigma_{5/2}$ of Cs and Ba and the asymmetry parameter $\beta_{5/2}$ for Cs have shown striking spin-orbit activated interchannel coupling near the thresholds of the $3d_{3/2}$ channels. The strongest effect on β occurs in the energy region where the mixing with the $3d_{3/2}$ channel produces a pronounced minimum of σ. In this case, the phase difference between the outgoing p and f waves plays the dominant role in the angular distribution of the photoelectrons.

We are grateful for the support of the BESSY staff during beam times. A good share of this work was thankfully funded by the Deutsche Forschungsgemeinschaft.

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