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### Ionic-configuration-interaction effects on Xe 5s-subshell photoionization processes

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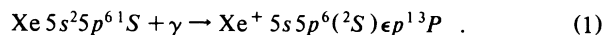
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Recent measurements of the 5s-subshell photoelectron angular distribution of xenon have found larger values for the asymmetry parameter  $\beta$  in the region of the 5s-subshell cross-section minimum than predicted by relativistic random-phase-approximation calculations. Final-state ionic configuration interaction is introduced as a possible explanation for this and other discrepancies.

#### I. INTRODUCTION

The spin-orbit interaction has been labeled<sup>1</sup> "a weak force with conspicuous effects." Among these have been the anomalous doublet intensity ratio,<sup>2</sup> the nonzero minimum in the photoionization cross section,<sup>3-5</sup> and the spin polarization of photoelectrons<sup>6-9</sup> in the alkalis as well as the unusual photoelectron angular distributions for s subshells in the alkalis and the rare gases.<sup>10</sup> All of these effects are conspicuous mainly near the photoelectron cross section minimum in the vicinity of threshold. Recent experimental measurements<sup>11,12</sup> of the photoelectron angular distribution of the 5s subshell in xenon indicate that the spin-orbit interaction is not the only weak interaction having conspicuous effects within such cross-section minima.

Within the electric dipole approximation, and considering only single configurations to describe initial and final states, the photoionization of the 5s subshell in xenon may be represented as the following process:



The transition amplitude for the  ${}^3P$  final state is nonzero only due to relativistic (mainly spin-orbit) interactions. Within the electric dipole approximation the photoelectron angular distribution asymmetry parameter is given by

$$\beta = (2 - r)/(1 + r) , \quad (2)$$

where  $r \equiv \sigma({}^3P)/\sigma({}^1P)$ . Here  $\sigma({}^1P)$  and  $\sigma({}^3P)$  are the photoionization cross sections for the  ${}^1P$  and  ${}^3P$  transitions. In the absence of relativistic interactions  $\sigma({}^3P)$  is zero and  $\beta$  is equal to 2. In practice,  $\sigma({}^3P) \ll \sigma({}^1P)$  so that  $\beta$  is close to 2 except in the region of the minimum in  $\sigma({}^1P)$ . [Note that  $\sigma({}^1P)$  never becomes exactly zero due to inter-channel interactions with photoelectrons from the 5p and 4d subshells, among others.] The dependence of  $\beta$  on photon energy in the region of the cross-section minimum in  $\sigma({}^1P)$

has been calculated in the relativistic random-phase approximation (RRPA) by Johnson and Cheng.<sup>13</sup> Their results, which incorporate effects of particle-hole interactions as well as relativistic interactions, agree beautifully with the first two experimental measurements<sup>14,15</sup> on either side of the cross-section minimum. More recent experimental measurements closer to the cross-section minimum<sup>11,12</sup> as well as at higher energies,<sup>12</sup> however, find a significantly higher value for  $\beta$  within the minimum and a somewhat lower value for  $\beta$  at higher energies than predicted theoretically.<sup>13</sup>

We point out here a usually weak interaction, besides the spin-orbit interaction, which has typically been ignored when describing photoionization processes theoretically, but which may have measurable effects when the dominant photoionization transition amplitude is small: final-state ionic configuration interaction. We propose that neglect of this interaction explains current discrepancies between theory and experiment not only for the  $\beta$  parameter but also for the partial cross section both near threshold and at higher energies.

Wang, Kim, Pratt, and Ron<sup>16</sup> have found that in photoionization of the 5s subshell of tin, interference of electric dipole and electric quadrupole transition amplitudes leads to corrections of greater than 5% to the pure electric dipole result for the differential cross section, especially in the neighborhood of cross-section minima. Such nonelectric dipole effects are probably just as significant for the xenon 5s subshell. However, final-state ionic-configuration-interaction effects influence not only the  $\beta$  parameter but also the cross sections and are important over a much wider energy range near threshold than electric quadrupole effects.

#### II. FINAL-STATE IONIC-CONFIGURATION-INTERACTION EFFECTS

The importance of configuration interaction in the final ionic state has been demonstrated in photoelectron spectroscopy.

copy experiments which find intense shakeup lines in addition to the expected main line.<sup>17</sup> In argon  $3s$ -subshell photoionization, the  $3s^23p^4(^1D)3d^2S$  excited configuration of the ion is produced with an intensity that is 15% of that of the main line.<sup>18</sup> The analogous  $5s^25p^4(^1D)5d^2S$  configuration in  $Xe^+$  is even more strongly mixed<sup>17,19-21</sup> with the usual ionic configuration  $5s^15p^6^2S$ . In a two-level theoretical treatment of process (1) the ionic Hamiltonian would be diagonalized to obtain two new eigenstates, each represented as a linear combination of the configurations  $5s5p^6^2S$  and  $5s^25p^4(^1D)5d^2S$ . The eigenstate with the lower energy would be a better representation for the ionic state than the single configuration  $5s5p^6^2S$ . One effect of such ionic configuration mixing in process (1) would be on the kinetic energy and wave function of the continuum electron, which would see both a lower ionization threshold and a less attractive ionic ground state. Another effect would be the modification of the  $5s$ - $5p$  intershell interaction. To the extent that the ratio  $\sigma(^3P)/\sigma(^1P)$  decreases in the neighborhood of the minimum in  $\sigma(^1P)$  as a result of these two effects, this configuration mixing might explain the discrepancy between theoretical and experimental values for  $\beta$ . In what follows we analyze the main discrepancies between the most detailed theoretical calculations for the  $Xe$   $5s$  subshell and experiment both for the  $\beta$  parameter and for the cross sections. We then discuss, using a two-level model, the main theoretical effects to be expected in a calculation including final-state ionic configuration interaction.

#### A. Discrepancies between present theory and experiment

Considering the strength of rearrangement processes in the  $Xe5s$  spectrum, it is by no means obvious that the RRPA should give good agreement with experimental  $Xe5s$ -subshell photoemission cross sections and  $\beta$  parameters. Experimentally, when one measures the variation of intensity of the  $5s$  photoelectron line with photon energy and emission angle one's measurements reflect the influence of all possible relaxation and correlation processes. The RRPA, on the other hand, refers to a single, unrelaxed  $5s$ -hole state, described by Dirac-Fock eigenvalues and wave functions calculated for the atomic ground-state configuration. It therefore employs a  $5s$  binding energy which is more than 4 eV too high and it does not describe satellite structure. Experimentally, however, there is important satellite structure, some of which arises from the  $5s$ -hole state and some from the  $5p$ -hole states. One may say that the RRPA describes the  $5s$  ionization without resolving the final ionic levels, so that at high energies above the  $5s$  threshold, the RRPA  $5s$ -subshell cross section should be compared with the *sum* of intensities of the experimental main  $5s$ -photoelectron line and its associated satellites and not just with that of the experimental main  $5s$  line. A theoretical estimate<sup>17,19</sup> gives a spectral weight of about 0.65 to the relaxed  $5s$  hole. The experimental cross section for the main  $5s$  line should thus lie well below the RRPA  $5s$  cross section at energies well above threshold.<sup>22</sup> It remains true also in the  $4d$ -resonance region in  $Xe$  (Ref. 23) and  $Ba$  metal.<sup>24,25</sup>

The situation becomes more complicated closer to the  $5s$  threshold because there the  $5p$  and  $5s$  excitation and ionization channels interact also via double excitation and ionization processes. It seems likely that part of the strong

“scatter” among the experimental points<sup>11,12</sup> and the nonsmoothness of the  $5s$  cross section and  $\beta$  parameter in the region from threshold through the minimum are in fact due to the double excitations, the satellite thresholds, the  $5p^2$  and  $5s5p$  double ionization thresholds, and associated dynamics. Near the  $5s$  threshold, therefore, one cannot, in principle, expect the RRPA to describe accurately even the  $5s$ -ionization cross section (i.e., the sum of the  $5s$  main line and the associated satellites).

At the minimum in both the  $\beta$  parameter and the cross section for the  $Xe5s$  subshell, the newest experiments<sup>11,12</sup> provide a sensitive test of theory. The  $\beta$ -parameter measurements give the triplet/singlet branching ratio (cf. Eq. 2): for  $\beta_{\min} \approx 1.4$ , we find  $r_{\max} \approx 0.25$ . The cross-section measurements<sup>22</sup> give the sum of the triplet and singlet cross sections

$$\sigma_{5s} = \sigma(^3P) + \sigma(^1P) , \quad (3)$$

where  $\sigma_{5s} \approx 0.05$  Mb at the minimum. Experiment thus predicts that  $\sigma_{\min}(^1P) \approx 0.04$  Mb and  $\sigma_{\min}(^3P) \approx 0.01$  Mb. Using Eqs. (2) and (3) to extract the RRPA values<sup>13</sup> for the  $^3P$  and  $^1P$  cross sections one finds that whereas the estimated  $^3P$  cross section in the minimum is comparable to the experimental one, the  $^1P$  cross section is roughly a factor of 6 too small. In making this comparison we have multiplied the RRPA cross sections by the spectral weight of 0.65 to take into account the  $\approx 35\%$  loss of the  $5s$ -subshell oscillator strength to satellite structure. If this is not done, then the calculated  $^3P$  cross section is roughly 1.5 times the experimental value and the calculated  $^1P$  cross section is a factor of 4 too small. In either case, the RRPA  $\beta$  parameters and cross section *at the minimum* are too small primarily because the  $^1P$  cross section is too small.

#### B. Two-level analysis of ionic-configuration-interaction effects

The framework for systematic<sup>26</sup> inclusion of relaxation effects in atomic photoionization calculations is quite well known.<sup>27,28</sup> However, only a few exploratory calculations make use of the many-body machinery to treat static and dynamic effects on the core-level shift and on the electron-hole interaction.<sup>27,29</sup> We discuss here qualitatively how the core-hole interaction process

$$5s^15p^6 \rightleftharpoons 5s^25^45d \text{ or } 5s \rightleftharpoons 5p^25d \quad (4)$$

influences the  $5s$  photoionization cross section. One should consider the entire  $5s^25p^4md$  discrete and continuum channel.<sup>17,19</sup> However, we limit ourselves to a two-level system, and ignore other effects such as double excitations  $5p^2mnp$  above the  $5s$  threshold.

##### 1. Effect on the $Xe5s$ state

The interaction in (4) leads to a  $5s$ -hole self-energy correction  $\Sigma_{5s}(E)$ , giving a dynamic correction  $E = E_s + \Sigma_{5s}(E)$  of the hole energy. The usual monopole (radial) relaxation process only contributes about 1 eV to the relaxation shift of a  $5s$  hole and is approximately compensated for by an opposite shift due to ground-state correlation effects.<sup>19</sup> However, the relaxation mechanism in (4) provides an energy shift of about 4 eV due to dipole (angular) relaxation,<sup>30</sup>

lowering the  $5\bar{s}$  binding energy from about 27.5 eV to about 23.5 eV. A photoelectron leaving the ion in the  $5\bar{s}$  ground state thus sees a less attractive potential than in the absence of configuration interaction.

Precisely because the  $5s$  photoionization process is critically dependent on the interaction with the  $5p$  and  $4d$  subshells, which largely determine the response of the  $5s$  subshell,<sup>31</sup> the  $5s$  ionization cross section is very sensitive to the position of the  $5\bar{s}$  threshold<sup>31</sup> and in particular its relation to the  $4\bar{d}$  and  $5\bar{p}$  thresholds. The Dirac-Fock  $5\bar{s}$  threshold is only about 15 eV away from the  $5\bar{p}$  threshold.<sup>32</sup> The relaxation effects described above reduce this energy difference by 4 eV. This reduction leads to a large increase of the  $5s$ -subshell  $^1P$  cross section in the threshold region, as demonstrated by Amusia and co-workers<sup>31,33</sup> in a nonrelativistic RPA calculation. Such an increase in  $\sigma(^1P)$  in the minimum would lead to a great improvement in the theoretical  $\beta$  parameter.

The interaction in (4) leads also to the reduction  $Z_{5\bar{s}}$  of

$$\text{Xe } 4d^{10}5s^25p^6\ ^1S_0 + \gamma \rightarrow \text{Xe}^+ 4d^95s^25p^6(^2D)\epsilon''f^{2S+1}P \quad (5a)$$

$$\rightarrow \text{Xe}^+ 4d^{10}5s^25p^5(^2P)\epsilon'd^{2S+1}P \quad (5b)$$

$$\rightarrow \text{Xe}^+ [C_15s5p^6(^2S) + C_25s^25p^4(^1D)5d(^2S)]\epsilon p^{2S+1}P \quad (5c)$$

Owing to final-state ionic configuration mixing  $|C_1| < 1$  and  $|C_2| \neq 0$  in channel (5c). The effect of this change on the interchannel coupling matrix elements is as follows: The Coulomb matrix element between channels (5a) and (5b) is unchanged. That between channels (5a) and (5c) is changed only slightly since  $|C_1| < 1$ , but otherwise the configuration preceded by  $C_2$  makes no contribution. The most important change is in the Coulomb matrix element between channels (5b) and (5c), which becomes

$$\begin{aligned} \langle (5b) \left| \sum_{i>j} \frac{1}{r_{ij}} \right| (5c) \rangle &= 2^{1/2} C_1 [\delta_{SO} 2R^1(5p\epsilon p; \epsilon'd5s)/3 - R^1(5p\epsilon p; 5s\epsilon'd)/3] \\ &+ 3^{-1/2} C_2 [2R^1(5d\epsilon p; 5p\epsilon'd)/3 + (-1)^S R^0(5d\epsilon p; \epsilon'd5p) + (-1)^S R^2(5p\epsilon p; \epsilon'd5p)/5] \quad (6) \end{aligned}$$

Here  $R^K(n_1l_1, n_2l_2; n'_1l'_1, n'_2l'_2)$  is the Slater radial integral. One sees that for a  $|C_2|$  value significantly greater than zero, the interaction between the  $5p$ - and  $5s$ -subshell channels (5b) and (5c) is completely altered. Furthermore, the interactions are quite different for singlet ( $S=0$ ) and triplet ( $S=1$ ) channels. Note that in a many-body perturbation theory or RPA calculation one does not usually diagonalize the configurations in (4) to obtain the weight factors  $C_1$  and  $C_2$  in Eqs. (5c) and (6). Rather one computes the relevant corrections order by order.

### C. Effect of $4p$ - and $4s$ -subshell channels

Finally, Fahlman, Carlson, and Krause<sup>12</sup> propose that omission of the  $4p$  and  $4s$  ionization channels might be partly responsible for the too large values of the RPA  $5s$  cross sections at energies well above threshold. The answer is most certainly negative: A local-density-based RPA calculation (LDRPA) (Refs. 25, 30, and 35) for atomic Ba including all channels involving the  $6s$ - $3d$  subshells shows that omission of the  $4p$ ,  $4s$ , and the  $3d$  subshells only increases the  $5s$  cross section by less than 3% in the  $4d$ -resonance region. A similar result must be true also for Xe and suggests that the spectral strength reduction factor  $Z_{5\bar{s}}$  is essential.

the intensity of the  $5s$ -photoelectron line,  $Z_{5\bar{s}} = (1 - \partial \text{Re} \Sigma_{5\bar{s}} / \partial E)^{-1}$ , and the appearance of a  $5p^25d$  satellite. The spectral strength factor  $Z_{5\bar{s}}$  is independent of energy and, since it is property of the ion, is *the same* for the singlet and triplet photoionization cross sections. For this reason it does not affect the  $\beta$  parameter, which depends only on the ratio of these cross sections, but it is essential for obtaining agreement with the experimental cross sections.  $Z_{5\bar{s}}$  is theoretically estimated to be 0.65.<sup>16,18</sup>

### 2. Effect on the $5s$ - $5p$ interchannel interaction

The major influence on the Xe  $5s$ -subshell photoionization cross section near threshold is the interaction with the  $5p$  subshell.<sup>34</sup> This interaction is significantly altered by the ionic configuration interaction in (4). Consider the most important channels involved in photoionization of the Xe  $5s$  subshell:

## IV. CONCLUSIONS

It seems clear that an RRPA calculation for the Xe  $5s$ -subshell cross section and  $\beta$  parameter should be carried out with use of the experimental  $5\bar{s}$  threshold. A systematic treatment requires also that the spectral strength factor  $Z_{5\bar{s}}$ , the corrections to the interchannel interaction, and the various dynamical effects not taken into account by a static approximation to the  $5\bar{s}$  self-energy be included. The nonrelativistic RPAE results suggest that in the threshold region the cancellation among these latter effects is almost complete<sup>28,31</sup> but this has to be confirmed by a relativistic calculation. As noted elsewhere,<sup>16</sup> the  $\beta$  parameter within the cross-section minimum may also be sensitive to electric quadrupole effects. The challenge of such a detailed calculation is to describe interaction effects not of the electric dipole, particle-hole type, which by now are well understood in photoionization processes involving closed-shell atoms.

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