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PHOTOIONIZATION CROSS SECTION MEASUREMENT IN 5p AND 5s SUBSHELLS OF EXCITED BARIUM ATOMS BETWEEN 40 eV AND 140 eV PHOTON ENERGY

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Résumé

Nous présentons ici la première mesure de sections efficaces de photoionisation en couches internes d’un atome préparé dans un état excité. Des atomes de baryum ont été portés dans les états excités 6s5d 1,3D à l’aide d’un laser continu, et ionisés en couche interne à l’aide du rayonnement synchrotron émis par ACO. Nous montrons que, dans ce cas particulier, le plus grand écrantage induit par l’électron 5d produit une diminution de l’énergie de liaison des électrons internes dans l’atome excité. Nous observons également une augmentation de l’interaction entre les voies 4d → εl et 5p → εl, 5s → εl au-dessus des seuils 4d.

Abstract

We present the first measurement of an inner-shell photoionization cross section in an excited atom. Barium atoms were laser-excited to the 6s5d 1,3D states and photoionized by synchrotron radiation from ACO. It is shown that the larger screening of the nuclear charge induced by the 5d electron leads to a decreasing of the binding energy of the inner electrons in the excited atom. We observe also an increasing of the interchannel interaction between the 4d → εl and 5p → εl, 5s → εl channels.

In the 80 eV–140 eV photon energy range, the photoabsorption spectrum of barium initially in its ground state (4d10 5s2 5p6 6s2 1S0 configuration) is known to be dominated by a broad and intense resonance above the 4d ionization thresholds [1], corresponding to a resonantly enhanced 4d → εf transition in the continuum, leaving only a weak oscillator strength to the discrete 4d → nl transitions [2]. Photoelectron studies using synchrotron radiation have shown in the atom [3,4] and in the solid [5] that this large resonance was also present in the 5p → εl and 5s → εl photoionization partial cross sections, produced via interchannel interaction with the 4d → εl channel. However photoabsorption spectra of Ba+ and mainly Ba+++ show a transfer of the oscillator strength from the continuum to the discrete part of the 4d spectrum, due to the decrease of the outer screening of the nuclear potential which induces the collapse of the 4f orbital [6]. Thus, barium represents a sensitive test of the screening, due to the outer electrons, of the effective potential experienced by inner electrons or by low-lying empty orbital.
Fig 1 : Photoelectron spectra recorded at 92.06 eV photon energy. Top panel: laser radiation off. The final states of Ba+ populated by photoionization of ground state Ba atoms are indicated above the corresponding electron peaks. Bottom panel: laser radiation on. New lines appear at lower binding energy than the previous ones. They are produced by photoionization of laser-excited atoms in the states 6s5d.

Very closely related studies have been recently reported on the 5p → nl discrete transitions in barium atoms excited in the states 6s 5d, realized by photoabsorption [7] and by resonant photoemission [8]. In this case, and like for the atom in its ground state, the giant resonance 5p → 5d which can be expected in view of the large overlap between the 5d and 5p orbitals, is in fact spread over a multitude of resonances, due to the collapse of the 5d lowlying orbital in the presence of the 5p hole.

We have previously reported results on photoionization cross section measurements in the 5d excited subshell of barium atoms prepared in the 6s 5d 1,3D states, between 40 eV and 140 eV photon energy [9]. Using the same experimental set-up, we present here results on 5p and 5s photoionization cross sections in the excited Ba atoms in the same energy range, covering discrete and continuum excitations of a 4d electron. This work constitutes the first measurement of an inner-shell photoionization cross section in an excited atom.

For this experiment, an effusive beam of Ba atoms was produced in a furnace heated at about 830 K, giving a typical density in the interaction volume of a few 10^{12} atoms/cm^3. The Ba beam was collinear with the synchrotron radiation (SR) beam produced by the ACO storage ring, monohromatized by a toroidal grating monochromator (line A61). A laser beam, from a CW ring dye laser, crossed the atomic beam at right angles in the interaction volume, and served to prepare the atoms in the excited states. Its wavelength was tuned and stabilized to the 6s^{2} 1S_{0} → 6s 6p 1P_{1} neutral Ba.
transition at 553.5 nm. The dye used was Rhodamine 110, providing laser power up to 600 mW. The 6s 6p 1P excited state rapidly decays to the 6s 5d 1,3D metastable states, in such a way that, under these experimental conditions, we have obtained up to 50% of the atoms prepared in these states. No signal from 6s 6p 1P atoms could be detected. The electrons emitted at the magic angle (54° 44') from the interaction volume between SR and excited Ba atoms were energy analyzed by a CMA, whose axis was colinear to SR beam. Since the atom density was high enough to destroy the alignment of the atoms induced by the polarization of the laser beam [10], this geometry allowed us to directly measure angularly integrated photoionization cross sections.

Figure 1 shows as an example photoelectron spectra recorded when the energy of the SR photons is adjusted to 92.06 eV. The spectrum on the top is obtained in the absence of the laser beam. We observe electrons lines produced by the photoionization in the outer and first inner subshells of barium atoms in the ground state: 6s (line at 87 eV kinetic energy), 5p (two lines around 68 eV corresponding to the 2 components 5p1/2 and 5p3/2) and 5s (line at 54 eV). Surnumerous lines are due to processes involving simultaneous excitation of at least one more electron to an empty orbital (satellite lines). The spectrum at the bottom is obtained with the laser beam irradiating the vapour. The lines due to photoionization of the ground state atoms are still present but reduced in intensity, since our vapour is constituted by atoms both in the ground and excited states. The intensity reduction of these lines allows us to measure the total population of atoms in excited states. Moreover, new lines appear at higher kinetic energy than the previous ones. They correspond to electrons produced by photoionization in the same subshells of atoms laser excited to the two 6s 5d 1,3D states, whose respective contribution is unresolved at this photon energy. The binding energy of the main lines we measure from such spectra, both in ground and excited states atoms, are listed in table 1. We observe that to promote a 6s electron to the 5d excited level results in a decrease of the binding energy of the inner electrons. This is in accordance with Wendin's calculation which has shown that the 5d orbital is more penetrating than the 6s one, thus screening more efficiently the nuclear potential experienced by the inner electrons [9].

TABLE 1: Binding energy of electron in the n1 subshell of Ba atoms in the ground state 6s2 1S and in the excited states 6s 5d 1,3D.

<table>
<thead>
<tr>
<th>nl</th>
<th>Ground State atoms 6s2 1S</th>
<th>Excited States Atoms 6s5d 1D</th>
<th>6s5d 3D</th>
</tr>
</thead>
<tbody>
<tr>
<td>5d</td>
<td>-</td>
<td>3.79 (4)</td>
<td>4.05 (4)</td>
</tr>
<tr>
<td>6s</td>
<td>5.19 (3)</td>
<td>4.51 (6)</td>
<td>4.72 (6)</td>
</tr>
<tr>
<td>5p1/2</td>
<td>24.75 (2)</td>
<td>~23.4</td>
<td>~22.0</td>
</tr>
<tr>
<td>5p3/2</td>
<td>22.72 (2)</td>
<td>20.5 (1)</td>
<td></td>
</tr>
<tr>
<td>5s</td>
<td>38.06 (6)</td>
<td>36.15 (15)</td>
<td></td>
</tr>
<tr>
<td>4d3/2</td>
<td>101.02 (6)</td>
<td>~99.0</td>
<td></td>
</tr>
<tr>
<td>4d5/2</td>
<td>98.41 (6)</td>
<td>96.4 (2)</td>
<td></td>
</tr>
</tbody>
</table>

a - The number in brackets gives the absolute uncertainty over the last number. When not indicated, the deconvolution from lines due to photoionization of ground state atoms was not possible.
Photoionization partial cross sections in the 5p and 5s subshells of laser-excited atoms are presented in figures 2 and 3 respectively, between 40 eV and 140 eV SR photon energy. Our measurements (points with error bars) are compared with the results of the one-electron calculation by Salzmann and Pratt (dashed line) [11]. Absolute values for our data are obtained like in [9], by normalization of the 5p photoionization cross section we measured for the ground state atom to the one calculated by Wendin using LDRPA. Our data do not include the contributions from all the satellite lines. For the 5p cross section we have measured the ratio sum of satellites to main line to be roughly constant and equal to 12% in this whole energy range, except in the 4d electron discrete excitations region we will discuss later. Outside the resonance region 85 eV - 120 eV, no differences for the 5p and 5s cross sections in the excited atom are measurable within our instrumental precision from those in the same subshell of the atom in its ground state [3]. This shows the weak coupling between the inner-shell electrons and the outer electron in what concerns continuum photoionization cross section. The same phenomenon has been observed for the 2p photoionization cross section between ground state 3s 2S1/2 sodium atoms and laser excited sodium atoms in the state 3p 2P3/2 (12). The presence of the large resonance around 100 eV, like for the Ba 5d cross section, proves that inter-channel coupling with the 4d \( \rightarrow \) 6l channels is still the dominating process. It is not described of course by the one-electron calculation.

We have studied in greater detail the most sensitive region between 90 eV and 102 eV of the discrete excitations of a 4d electron in the excited atom. The dominant photoionization processes can be described as:

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Fig 2 : Photoionization cross section in the 5p subshell of excited Ba atoms in the states 6s 5d \( \stackrel{1}{\overline{\ell}} \) 2D. This work. ----

Fig 3 : Photoionization cross section in the 5s subshell of excited Ba atoms in the states 6s 5d \( \stackrel{1}{\overline{\ell}} \) 2D. Same symbols as in fig. 2.
Resonant (autoionization, path (a,b)) and direct single ionization (path (c)) have the same order of magnitude. Autoionization to Ba^{n+} final states (n = 2,3), which is supposed to be strong, is not observable with our apparatus. Nevertheless, the comparison of the total yield of 4d Auger electrons and 4d photoelectrons we have observed for the ground state atom suggests that desexcitation of a 4d hole leads to a minimum ratio Ba^{n+}/Ba^{+} of 0.6. We have measured the excitation functions of all the Ba^{+} exit channels between 90 eV and the 4d thresholds. The main Ba^{+} exit channel is Ba^{+} 4d^{10} 5s^{2} 5p^{6} 6s 5d nl e', due to the strong overlap between 4d and 5p orbitals, producing the satellite structure observed between 40 eV and 50 eV binding energy on the spectra of the figure 1. We only present here (figure 4), the excitation functions for the sum of all the Ba^{+} exit channels, except the Ba^{+} 4d → e1...
channels above the 4d thresholds, for ground state atoms (middle panel) and excited states atoms (bottom panel). Photoabsorption spectrum recorded by Ederer et al [13] for the ground state Ba atoms is also reported in the top panel. Taking account of our lower resolution, it compares quite well with our excitation function for the ground state atoms, despite we do not include contribution from the Ba$^{n+}$ ($n>1$) exit channels. For excited atoms, the number of resonances is spread out and multiplied because of the greater number of allowed couplings, as observed in the 5p electron excitations region [7,8]. The two main resonances are shifted to lower energy by about the energy brought by the laser in the excited atoms. But the more striking phenomenon, as compared to the ground state atoms, is an increase in intensity of the large resonance in the continuum above the main 4d thresholds. So, to promote an outer electron to the 5d excited level seems to strengthen the interaction between the 5p, 5s $\rightarrow \epsilon_1$ and 4d $\rightarrow \epsilon_1$ channels.

REFERENCES