Photoionization of excited barium $6s6p\ ^1P_1$

V. Lange, U. Eichmann, and W. Sandner
Fakultät für Physik, Universität Freiburg, D-7800 Freiburg, Germany
(Received 31 January 1991)

To clarify some essential discrepancies between experimental and recent theoretical studies of the photoionization cross section of excited $6s6p\ ^1P_1$ barium, we present experimental data of the relative photoionization cross section of the $6s6p\ ^1P_1$ state of barium in the energy range between the Ba^+ $6s$ and Ba^+ $5d_{3/2}$ thresholds.

PACS number(s): 32.80.Fb, 32.30.Jc, 32.80.Rm

INTRODUCTION

Photoionization cross sections of alkaline-earth-element atoms have been measured experimentally in the past using either synchrotron radiation or, more recently, laser radition. Theoretically, much progress has been achieved through the development of variational eigenchannel R-matrix calculations in combination with multichannel quantum defect theory (MQDT) [1], and the conventional approach of the R-matrix formalism [2]. In cases of photoionization out of the ground state or highly excited Rydberg states, theory gives, in general, good agreement with experimental data, not only for the total cross section but also for the differential cross section and branching ratios of the photoelectrons [1–6]. The situation is somewhat different for low-lying excited states. In the alkaline-earth-element atom barium, cross-section data for the photoionization out of excited states $5d6l$ and $6s7p$ have been experimentally studied by Camus *et al.* [7] but only empirically parametrized with the help of MQDT by Aymar, Camus, and Himdy [8]. Only the $6s6p\ ^1p_1$ state in barium is, accidentally, a singular case: on the one hand, there exists only a small data set of absolute experimental photoionization cross sections at a few discrete excess energies between the Ba^+ $6s$ and Ba^+ $5d_{3/2}$ thresholds [9,10]. On the other hand, R-matrix calculations using both the variational approach with a jj-coupled basis set [11] and the conventional R-matrix approach [12] have been applied in this energy range and lead to different results in some prominent features of the spectra. Comparison of these calculations with the existing experimental data indicates that the variational R-matrix calculations, which give good agreement for ground-state and Rydberg-state photoionization, are in rather poor agreement with the experiment.

This discrepancy and the need for more complete experimental data stimulated us to investigate the photoionization cross section out of the $6s6p\ ^1P_1$ state in barium over the complete energy range between the Ba^+ $6s$ and Ba^+ $5d_{3/2}$ thresholds. Since we are not able to determine absolute cross sections, we took special care to achieve good energy calibration of the spectra, and reliable relative measurements of cross section.

EXPERIMENT

In the present experiment we used two pulsed dye lasers to carry out the photoionization. The first one, a Littman-type dye laser [13], was tuned to 553.7 nm to excite the $6s6p\ ^1P_1$ state in barium. The second dye laser was a commercial Lambda Physik FL3002 dye laser with 0.2 cm^{-1} bandwidth, operated at a power of less than 0.5 mJ. Both dye lasers were only slightly focused to a diameter of about 1 mm on the excitation region to avoid multiphoton processes due to high power densities. The second dye laser was tuned between 418 and 345.76 nm (wavelength in air) to cover the whole energy range between the Ba^+6s limit and the Ba^+ $5d_{3/2}$ threshold. Four different dyes were used for the energy range under study, providing sufficient energy overlap of different laser scans. Both lasers were linearly polarized, either parallel, giving $\Delta m=0$ transitions only (m is the magnetic quantum number) for the excitation of $J=0,2$ final states, or perpendicularly ($|\Delta m|=1$ transition), where final states with $J=0$ are excluded. The data were taken with both polarizations in the whole energy range. The second dye laser was delayed by a few nanoseconds with respect to the first one in order to suppress excitation with the second dye laser and photoionization with the first one.

In the vacuum chamber (background pressure $\sim 1.5\times 10^{-9}$ Pa), which has been described in detail elsewhere [14], the laser beams crossed perpendicularly an effusive atomic beam between two field plates, which were grounded during the excitation process. The atomic density in the excitation region was about 10^9 atoms cm^{-3}. After the second laser had fired, a field pulse of about 300 V was applied to one of the field plates to drive the ions out of the excitation region towards the microchannel plate ion detector. The analog signal of this detector was digitized with a gated analog-to-digital converter and stored in a computer for further evaluation. The photoionization spectra as a function of the laser wavelength were measured simultaneously (a) with the laser power, using a calibrated Molectron detector system (J3S-10UV) to obtain the relative efficiency curves of each dye, (b) with the optogalvanic Ar I excitation spectrum in

a hollow cathode, which is known to an accuracy of better than 0.2 cm^{-1}, for absolute calibration of the energy scale, and (c) with the fringes of an étalon with a free spectral range of 0.82 cm^{-1}, which serves as a relative frequency measurement. The measurement of the laser power was necessary to normalize the amplitude of the ion signal, while the density of the atomic beam was kept constant during the measurement.

EXPERIMENTAL RESULTS AND DISCUSSION

Photoionization spectra of the $6s6p\ ^1p_1$ state of barium (corrected for laser-power fluctuations) are given in Figs. 1(a)–1(e) for parallel and in Figs. 2(a)–2(e) for perpendicular polarization of the two laser beams. The energy range corresponds to wavelengths from 418.0 to 345.76 nm for the second laser, covering the range from just below the Ba^+ 6s threshold to slightly above the $5d_{3/2}$ limit of Ba^+. Note that the displayed energy scale of every diagram is very different, but that the scale of the y axis is always the same.

When comparing the data for both polarizations we find two striking differences. First, the broad and strong enhancement of the cross section around 374 nm for parallel polarizaiton [Fig. 1(a)] disappears with perpendicular polarization [Fig. 2(a)]. Consequently, this broad resonance must have $J=0$, in agreement with predictions of Aymar, Camus, and Himdy [15], who labeled it $6p^2\ ^1S_0$.

Second, focusing on the end of the measured energy range, just below the Ba^+ $5d_{3/2}$ threshold [Figs. 1(c),

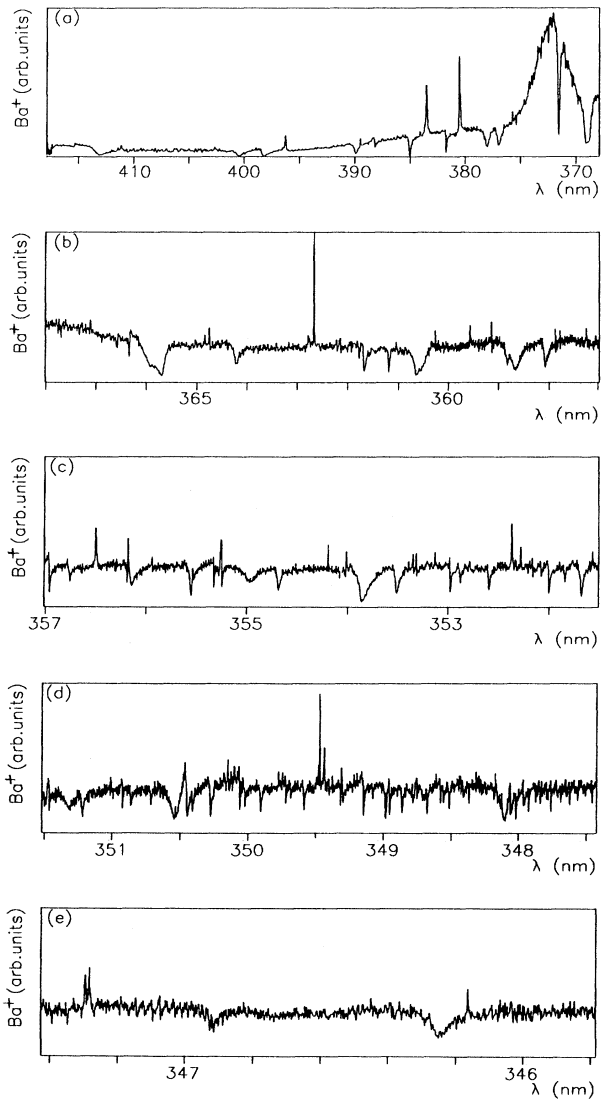

FIG. 1. Relative cross section (normalized to the laser power) for photoionization of the $6s6p\ ^1P_1$ state in barium as a function of the wavelength (in air) of the second laser in the energy range between the Ba^+ 6s and Ba^+ $5d_{3/2}$ thresholds, for parallel polarization of the lasers.

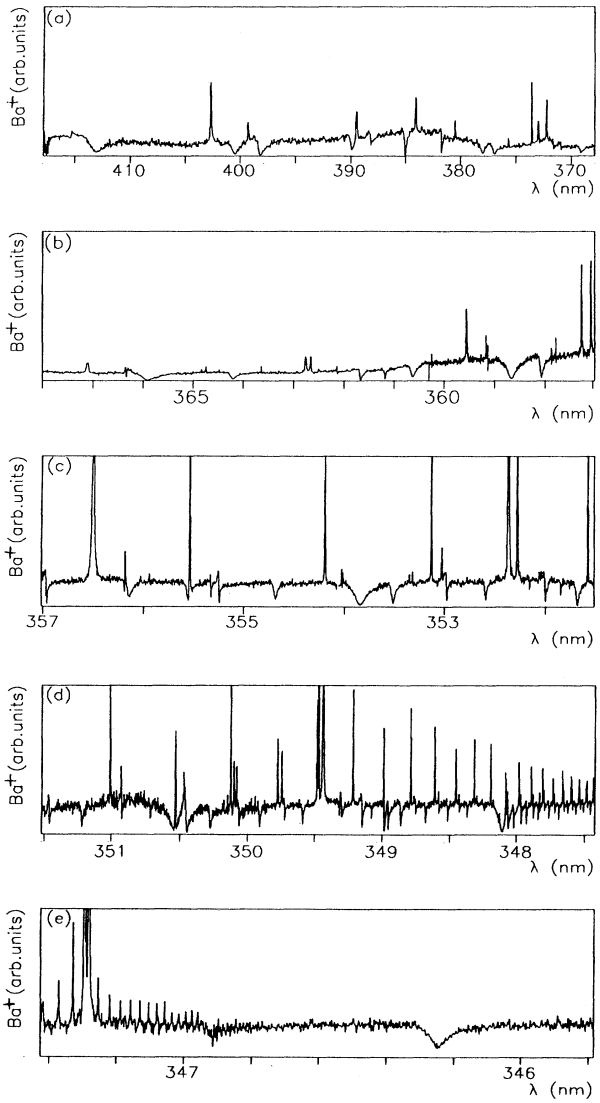

FIG. 2. Same as in Fig. 1 but with perpendicular polarization of the lasers.

1(d), 2(c) and 2(d)], we identify a Rydberg series of narrow resonances with a high Fano q parameter, which is absent in case of parallel polarization, indicating a total angular momentum $J=1$. Furthermore, we observe two Rydberg series of window resonances (Fano q parameter $q\simeq 0$) for both polarizations. From the fact that these two series appear for both polarizations, we can conclude that they have $J=2$. All three series converge to the Ba^+ $5d_{3/2}$ ionization limit. The effective quantum numbers $\nu_{5d_{3/2}}(\mathrm{mod}1)$ of the two series of window resonances are $\simeq 0.44$ and $\simeq 0.86$ with respect to the Ba^+ $5d_{3/2}$ threshold, while the series of narrow resonances has an effective quantum number $\nu_{5d_{3/2}}(\mathrm{mod}1)\simeq 0.25$. Comparison of our results with the analysis from Aymar, Camus, and Himdy [8] enabled us to identify the series of narrow resonances with $5d_{3/2}nd_{3/2}$ $(J=1)$ and the two series of window resonances with the $5d_{3/2}nd_{3/2}$ $(J=2)$ and the $5d_{3/2}ns$ $(J=2)$ Rydberg series. We remark that the width of the $5d_{3/2}nd_{3/2}$ $(J=1)$ series is very small, for states with $n\simeq 17$ we observe a width of roughly 0.5 cm^{-1}.

To compare our data with the controversial theoretical cross-section data we shall concentrate on the energy range just above the Ba^+ $6s$ ionization limit around 415 nm. In particular we observe the first minimum [window resonance, which is denoted $5d_{3/2}9s$ $(J=2)$ [7,8]] in the photoionization cross section at 413.2 nm, in excellent agreement with the R-matrix calculations of Greene and Theodosiou [11], while both the calculations of Bartschat and McLaughlin [12] and the experiment of Kallenbach, Kock and Zierer [10] show a minimum around 412 nm. We are rather confident about the accuracy (<0.2 cm^{-1}) of our measured absolute energy positions; note also that this particular state belongs to the identified series of window resonances with $\nu_{\mathrm{eff}}(\mathrm{mod}1)=0.85$ and has therefore the same effective quantum number modulo 1. The next test of the theory is the shape and the energy position of two small resonances in the photoionization cross section at 415.3 nm [denoted $5d_{5/2}7d_{5/2}$ by Aymar, Camus, and Himdy [8]] and at 411.2 nm [denoted $5d_{5/2}7d_{5/2}$ $(J=2)$ in Ref. [8]]. They are present in the calculation of Greene and Theodosiou [11], but not in the calculation of Bartschat and McLaughlin [12]. Because of the large error bars in the experimental results of Kallenbach, Kock, and Zierer, [10] no information about these resonances can be obtained from their data. Due to the selection rules the resonance at 411.2 nm is only visible with parallel polarized light (Fig. 1). While the energy positions of the resonances agree well with the calculations of Greene and Theodosiou [11] the q parameter of the resonance at 415.3 nm has obviously the wrong sign. The two strong resonances appearing at 415.8 and 409.5 nm in the calculation of Bartschat and Mclaughlin [12] were not observed in the experiment.

Comparison of the energy range around the $6p^2\,{}^1S_0$ resonance around 374 nm reveals, that the calculation of Greene and Theodosiou [11] predicts the envelope of the resonance and its position quite well, while the detailed structure seems to be composed of a superposition of the experimental spectra of both polarizations. Inspecting the energy range around 354 nm, given in Figs. 1(c) and 2(c), with Fig. 2 of Greene and Theodosiou [11] indicates that the energy position of some features agrees with our experimental data, but significant differences in the shape (q parameter) of the resonances prevail. For even higher excess photon energies, reaching just below the Ba^+ $5d_{3/2}$ limit, no comparison can be made due to the finite resolution of the calculations of Ref. [11].

SUMMARY

In this experiment we have performed a detailed study of the photoionization cross section of excited $6s6p\,{}^1P_1$ barium in the energy range between the Ba^+ $6s$ and Ba^+ $5d_{3/2}$ ionization limits. We conclude that the theoretical calculations of Greene and Theodousiou [11] come close to the measured relative cross section concerning the energetical position of the prominent features of the spectra, but not in every detail of the shape of these resonance profiles. Therefore, new calculations, taking also into account the polarization of the lasers, would help to clarify the observed discrepancies.

ACKNOWLEDGMENTS

The authors thank C. E. Theodosiou for some stimulating discussions. The work was supported by the Deutsche Forschungsgemeinschaft (Sonderforschungsbereich 276).

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