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Channel interaction of Ba 6pns (J=1) autoionizing series*

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The channel interactions between the Ba 6pnl (J=1) autoionizing series with different l and n have been observed by means of unsaturation and saturation excitation methods. The observed interactions were interpreted with the spectral density calculated from the multichannel quantum defect theory, the results of which are quite satisfactory.

Keywords: channel interaction, autoionizing series, MQDT, ICE
PACC: 3260, 3280

1. Introduction

The channel interaction of autoionizing series in an alkaline-earth atom reveals the perturbation between the valence electrons of the alkaline-earth atom. Thus, analysis of channel interaction is a crucial method for interpreting this physical mechanism. Generally speaking, there are two types of channel interactions in the autoionizing series that embed in several continua.[1] The first type is the interaction of autoionizing states with the continua, which results in a larger width of an autoionizing state. The second type is the interaction among the autoionizing states with different n and l, leading to complex structures on the spectra of autoionizing states. In this paper we focus on the second type of channel interaction for the Ba 6pns (J=1) series. For the Ba $6p_{3/2}ns$ (J=1) series, Cooke and Cromer[2] have observed the interaction of the $6p_{3/2}12s$ state with the higher n members of $6p_{1/2}nl$ (J=1) series. Recently, Lu *et al*[3] observed the interactions of the lowest $6p_{3/2}ns$ states (n=8–11) with the $6p_{1/2}ns$ and $6p_{1/2}nd$ states. However, all of the above-mentioned $6p_{3/2}ns$ states are located below the $6p_{1/2}$ ionization threshold. Here, we report on the first observation on the interactions of $6p_{3/2}nl$ states located above the $6p_{1/2}$ ionization threshold. Furthermore, since the channel interaction is a mutual effect, the higher n members of the $6p_{1/2}ns$ series will reflect the impact from the lowest n $6p_{3/2}ns$ states. Therefore, in order to explore these interactions we have made a first attempt to excite the saturation spectra of the $6p_{1/2}ns$ states. On the theoretical side, the multichannel quantum defect theory (MQDT) combined with R-matrix calculation has played an important role in studying autoionizing state, and has had great success in several aspects, such as the cross section of autoionizing states, branching ratio, angular distribution of electrons ejected from autoionizing states[4–6] and the Stark structure.[7,8] Here, we utilize a 13-channel MQDT model to analyse the channel interactions of 6pns states by calculating their spectral density or channel mixing coefficient. The agreement between the experimental and theoretical results is quite satisfactory.

2. Experimental observations

The experimental apparatus is similar to those described in the previous works,[9,10] so only the main features relevant to this experiment are described here. The apparatus includes several parts for electrothermal atomization, laser resonance excitation and ion signal collection. The Ba atom beam is extracted from a resistively heated oven, and interacts perpendicularly with the laser beams from the three Nd:YAG laser-pumped dye lasers, which typically have an energy of 100μJ and a linewidth of 0.5 cm^{-1}. The atom in the 6pns autoionizing state quickly decays into an electron and an ion. In order to collect the ion signal, a 100V pulsed electric field

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is applied to the two parallel plates. Finally, the ion signal is detected by a microchannel plate (MCP) detector.

In this experiment we used two efficient experimental methods for observing the channel interaction of autoionizing series. The first method is to excite the normal spectrum of the autoionizing state with the lower laser power of the third laser. In order to obtain the 6pns spectrum, the three dye lasers are all linearly polarized in the same direction so that Δm_J=0 and $|\Delta J|$=1 hold for each step of the transition. Particularly, the first two lasers excite the Ba atom from the $6s^2\ ^1S_0$ ground state to the different 6s$ns\ ^1S_0$ Rydberg states via the intermediate 6s6p $^1P_1$ state, while the third laser excites the other valence electron from the 6s to the 6p ionic state. In the third step of excitation, if the power of the third laser is kept at a proper value, the Rydberg electron will not transit and will remain a spectator, which performs the so-called isolated core excitation (ICE) scheme. In most cases, the ICE spectrum exhibits a nearly Lorentzian profile. However, the spectrum of the interacting autoionizing state will exhibit some complex structures. Thus, this method enables us to observe the channel interaction of the autoionizing series. The second method we used is to examine the saturation spectrum obtained with the high power of the third laser. In other words, to saturate the 6s$\rightarrow$6p$_{1/2}$ transition, we deliberately increased the power of the third laser about 50 times. In this case, the energy absorbed by the Rydberg electron can no longer be neglected due to the high power of the third laser, resulting in the appearance of many satellite peaks in the wings of the main Lorentzian profile. Since the features of satellite peaks in the saturation spectrum can provide important information about the autoionizing series, the saturation excitation method has been an efficient tool for observing the channel interactions.[11]

In this paper, the spectrum of the 6p$_{3/2}$15s state and the saturation spectra of the 6p$_{1/2}$19s and 6p$_{1/2}$31s states are taken as examples to illustrate how to use the two methods mentioned to investigate the channel interactions. To demonstrate the first method, an example of many experimental spectra is shown in Fig.1(a), which is obtained by driving the 6s15s^{1}S$_0$ $\rightarrow$6p$_{3/2}$15s transition. This non-Lorentzian profile indicates that the 6p$_{3/2}$15s state is affected by the other 6pnl members. As is known, for the odd-parity J=1 series there are five channels converging to the 6p$_{1/2}$ and 6p$_{3/2}$ thresholds, namely the 6p$_{1/2}n$s$_{1/2}$, 6p$_{1/2}n$d$_{3/2}$, 6p$_{3/2}n$s$_{1/2}$, 6p$_{3/2}n$d$_{3/2}$ and 6p$_{3/2}n$d$_{5/2}$ channels. Since the 6p$_{3/2}$15s state lies above the 6p$_{1/2}$ threshold, only the 6p$_{3/2}n$d$_{3/2}$ and 6p$_{3/2}n$d$_{5/2}$ channels may generate the complex structures on the spectrum of 6p$_{3/2}$15s state. With the second method, an example of many observed saturation spectra is shown in Fig.2(a), which is obtained by exciting the 6s19s $^1S_0$ Rydberg state with higher power. This saturation spectrum covers many 6p$_{1/2}n$s states with n=16–27. In order to identify these 6p$_{1/2}n$s states, the unsaturated spectrum of the 6p$_{1/2}$19s state is taken as a reference, as shown in Fig.2(b). The fact that there is no additional feature on each n peak in Fig.2(a) indicates that the 6p$_{1/2}n$s series does not interact with the 6p$_{1/2}n$d series. Hence, there are only the interactions between the autoionizing series converging to the different thresholds, i.e., the interaction between the higher n members of the 6p$_{1/2}n$s series and the lowest n members of the 6p$_{3/2}n$s states. Figure 2(a) shows that the saturation spectrum of the 6p$_{1/2}$31s state is superimposed on a base signal, which is due to the interactions of the higher n members of the 6p$_{1/2}n$s series with the 6p$_{3/2}$12s state. This is clear evidence that the 6p$_{3/2}$12s state indeed affects the higher n members of the 6p$_{1/2}n$s series.

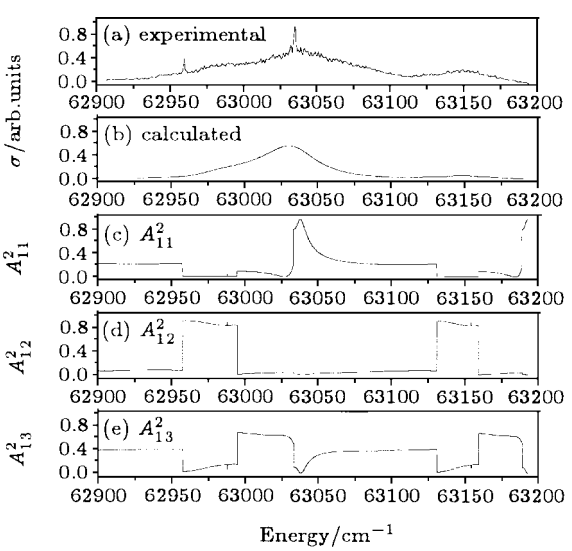


Fig.1. Interpretation of the interactions of the 6p$_{3/2}$15s state with the 6p$_{3/2}n$d $_{3/2}$ and 6p$_{3/2}n$d $_{5/2}$ states. (a) The experimental spectrum of the 6p$_{3/2}$15s state. The sharp peaks in the observed spectrum are the two-photon-transition signals from the third laser, and can serve as frequency markers. (b) The calculated spectrum of the 6p$_{3/2}$15s state from a 13-channel MQDT model. (c), (d) The spectra of A_{12}^2 and A_{13}^2, corresponding to 6p$_{3/2}n$d$_{3/2}$ and 6p$_{3/2}n$d$_{3/2}$ channels, respectively.

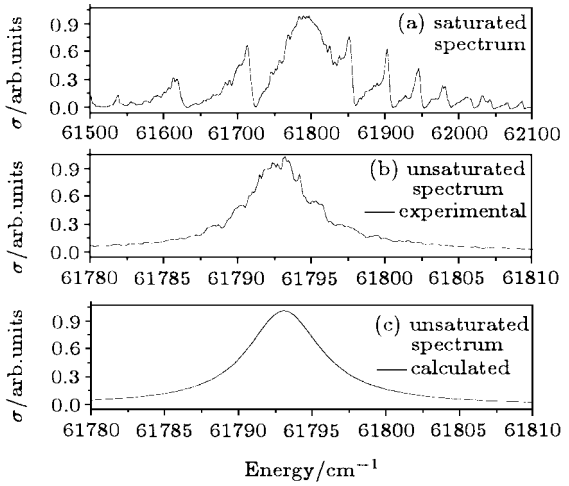


Fig.2. The saturation and unsaturation spectra of the $6p_{1/2}19s$ state. (a) The saturation spectrum of the $6p_{1/2}19s$ state. The spectrum covers many $6p_{1/2}ns$ states with n=16 to 27. (b), (c) The experimental and calculated $6p_{1/2}19s$ unsaturation spectra. A comparison between them shows that the experimental results are in good agreement with the results from MQDT.

3. Theoretical analysis

Further investigation of interactions motivates us to calculate the channel mixing coefficients of the five channels converging to the $6p_{1/2}$ and $6p_{3/2}$ thresholds. The channel mixing coefficient and cross section for the Ba J=1 autoionizing state can be calculated with a 13-channel MQDT model, whose parameters are similar to those in Ref.[12]. Since the MQDT is well documented, we only describe the main equations to be used in our study. There are 13 channels existing for the J=1 autoionizing series, namely the $6s_{1/2}np_{1/2}$, $6s_{1/2}np_{3/2}$, $5d_{3/2}np_{1/2}$, $5d_{3/2}np_{3/2}$, $5d_{3/2}nf_{5/2}$, $5d_{5/2}np_{3/2}$, $5d_{5/2}nf_{5/2}$, $5d_{5/2}nf_{7/2}$, $6p_{1/2}ns_{1/2}$, $6p_{1/2}nd_{3/2}$, $6p_{3/2}ns_{1/2}$, $6p_{3/2}nd_{3/2}$ and $6p_{3/2}nd_{5/2}$, which converge to the $6s_{1/2}$, $5d_{3/2}$, $5d_{5/2}$, $6p_{1/2}$ and $6p_{3/2}$ thresholds. These dissociation channels are labelled as i=1 to 13. In MQDT, the spectral density[13–15] or square of channel mixing coefficient corresponding to the ith dissociation channel is determined by

$$A_i^2 = \sum_\rho A_i^2 \rho, \tag{1}$$

where ρ refers to the collision eigenchannel. $A_{i\rho}$ are the coefficients in the wavefunction of the collision eigenchannel, and can be expressed as

$$A_{i\rho} = \frac{1}{N_\rho} \sum_\alpha U_{i\alpha} \cos[\pi(\nu_i + \mu_\alpha)] B_\alpha^{(\rho)}, \tag{2}$$

where ν_i is the effective quantum number for a closed channel, and is replaced with $-\tau_\rho$ for an open channel. N_ρ is the normalization constant. $B_\alpha^{(\rho)}$ is the coefficient of wavefunction φ_α in the αth close-coupled eigenchannel, which can be obtained by solving the MQDT equations.[16] Furthermore, the partial cross section σ_{J_c} to a particular final ionic state Ba (J_c) and the total cross section σ are

$$\sigma_{J_c} = \frac{4}{3}\pi^2 \alpha \hbar\omega_3 \sum_i \left| \sum_\rho D_\rho \exp(\mathrm{i}\pi\tau_\rho) A_{i\rho} \right|^2 \tag{3}$$

and

$$\sigma = \sum_{J_c} \sigma_{J_c}. \tag{4}$$

Here $\hbar\omega_3$ is the photon energy of the third laser, and D_ρ is the corresponding reduced transition matrix element. The first sum in equation (3) is only taken over the open dissociation channels corresponding to the ionic states.

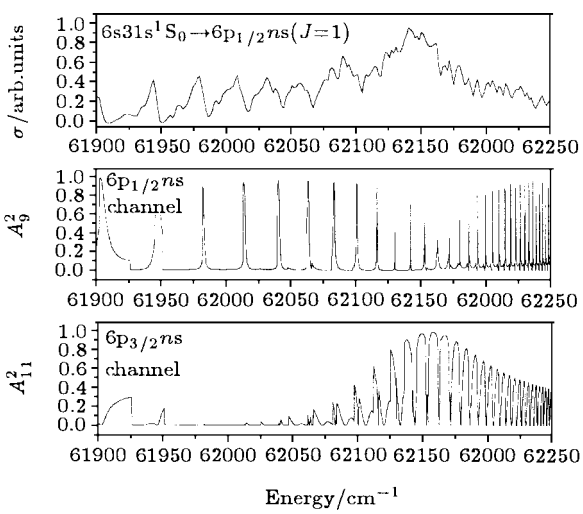


Fig.3. Manifestation of the interactions between the higher n members of the $6p_{1/2}ns$ series and the $6p_{3/2}12s$ state. (a) The saturation spectrum of the $6p_{1/2}31s$ state. The base signal is due to the admixture of the $6p_{3/2}12s$ state. (b), (c) The spectra of A_9^2 and A_{11}^2, corresponding to $6p_{1/2}ns_{1/2}$ and $6p_{3/2}ns_{1/2}$ channels, respectively.

Two examples of many calculated $6p_{1/2}ns$ and $6p_{3/2}ns$ spectra are shown in Figs.1(b) and 2(c), respectively. The results of comparison between the theoretical and experimental spectra show that the theory is in good agreement with the experiment. Although the combination of R-matrix with the MQDT has had great success in the Ba atom,[3–6] there has been little concentration on the analysis of channel interaction. With the correct MQDT parameters we take a further step to study the channel interactions of the $6pns$ series by calculating the corresponding spectral density A_i^2. The calculated results of A_i^2(i=9–13)

show that the interactions of the $6p_{3/2}15s$ state with the $6p_{1/2}ns$ and $6p_{1/2}nd_{3/2}$ channels can be neglected, and interactions with the $6p_{3/2}nd_{3/2}$ and $6p_{3/2}nd_{5/2}$ channels prevail, as shown in Figs.1(c)–(e). Comparing these figures with either experimental or calculated spectra, one may conclude that the main peak in the $6p_{3/2}15s$ spectrum is due to the contribution from the $6p_{3/2}ns$ channel, while the contributions from the $6p_{3/2}nd_{3/2}$ and $6p_{3/2}nd_{5/2}$ channels lead to the subsidiary peaks on both sides. With the knowledge of the $6p_{3/2}nd$ and $6p_{3/2}ns$ series, the peak on the red side can be attributed to the admixtures of $6p_{3/2}14d_{3/2}$ and $6p_{3/2}14d_{5/2}$ states, and those on the blue side to the mixtures of $6p_{3/2}15d_{3/2}$ and $6p_{3/2}15d_{5/2}$ states. Similar observations and calculations also have been carried out for the $6p_{3/2}13s$ and $6p_{3/2}14s$ states, the results of which are satisfactory to the same degree. For the $6p_{1/2}ns$ series, the interaction observed in Fig.3(a) can be confirmed with the spectral density A_9^2 and A_{11}^2, as shown in Figs.3(b) and (c). With these results one may conclude that the base signal is due to the admixture of the $6p_{3/2}12s$ state. Furthermore, both the observed spectrum and the theoretical results demonstrate that the most strong admixture occurs in the $6p_{1/2}31s$ state. The spectrum of A_{10}^2 also confirm that the $6p_{1/2}ns$ series hardly interacts with the $6p_{1/2}nd_{3/2}$ series, which is evident in Fig.2(a).

4. Conclusions

In summary, we have employed an ICE method to study the $6p_{3/2}ns$ states above the $6p_{1/2}$ threshold, whose interactions with the $6p_{3/2}nd$ states are evident in the observed spectra. No interaction between the $6p_{1/2}ns$ series and the $6p_{1/2}nd$ state is observed. Interaction between the higher n members of the $6p_{1/2}ns$ series and the $6p_{3/2}12s$ state is observed using the saturation excitation method. All the observed channel interactions are well interpreted with the MQDT. A systematic study of the spectroscopic properties of the Ba 6pns (J=0,1) autoionizing series, including energy positions and autoionization rates, is in progress and will be presented elsewhere.

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