

# Summary and Outlook

In this dissertation, the photoexcitation process and the subsequent autoionization decay of doubly excited helium were studied experimentally and theoretically in a broad photon energy region, where the spectra exhibit a characteristic change from strong electron correlation to quantum chaos.

On the experimental side, two different types of experiments were performed. The first type was the experimental study of the total cross sections (TCS) by measuring the total ion yield using a gas cell. With a high-resolution monochromators at the third generation synchrotron radiation sources the present studies of the TCSs were first extended to the single ionization threshold (SIT),  $I_{14}$ , of  $\text{He}^+$ . These new data agree remarkably well with the results of very recent state-of-the-art complex-rotation calculations by Delande [27], which could be performed up to the SIT  $I_{17}$ . The present experimental spectra together with these confirmed theoretical results were employed to study signatures of quantum chaos in doubly excited helium in the region very close to double-ionization threshold.

The experimental spectra below each threshold can be described well by a describing fit using only few number (about 15) of resonances, where most of these resonances belong to the principal Rydberg series. Since there are normally several hundred of resonances in the region chosen for the fits, the resonances are strongly overlapping, a situation that in principle results in so-called Ericson fluctuations [26], i.e. one of the features of quantum chaos. On the basis of theoretical data, we found that the angular correlation quantum numbers  $K$  for the principal Rydberg series are still good quantum numbers at least up to the SIT  $I_{17}$ . Since the resonances with  $K = N - 2$  carry most of the intensity of the cross sections, we can understand why the Ericson fluctuations were not observed in doubly excited helium although the spectra studied are in the Ericson regime, i.e. they are strongly overlapping. The present conclusions demonstrate that the conditions of comparable intensities for the resonances are needed to observe Ericson fluctuations. It is not reliable to prove the Ericson fluctuations only by the autocorrelation function with a Lorentzian form. The calculated  $K$  values are found to be good quantum numbers for resonances with  $K \rightarrow K_{max}$  and  $K \rightarrow K_{min}$ , but not for  $K \rightarrow 0$ . These can be well understood by the classical collinear  $eZe$  and  $Zee$  configurations. On the basis of the present calculated  $K$  values for the principal Rydberg series, we analyzed the angular correlation mechanisms below and above the double-ionization threshold, which shows the photon double-ionization process is described by the same mechanism with photon single ionization, particularly for the regions very close to double-ionization threshold.

The assignment of the spectra was done for the first time up to  $I_{14}$  in this dissertation.

Statistical properties of the energy levels, linewidths, and Fano  $q$  parameters can also demonstrate quantum signatures of chaos. These statistical properties were studied on the basis of theoretical data [27], since only a small fraction of the resonances can be observed experimentally. In order to derive different aspects of quantum chaos different kinds of the nearest-neighbor spacings (NNS) were performed. First, a global analysis of the NNSs, i.e. all resonances were taken into account without further restriction. The distribution derived from this analysis results in a Poisson distribution up to the SIT  $I_{16}$ . This indicates that there are still independent subsystems, which can be identified by good quantum numbers like  $K$ . These studies prove the transition region from integrability to quantum chaos to be much larger than expected before, since the quantum number  $K$  dissolves rather slowly. Full chaos in  $^1P^o$  doubly excited helium may occur only at the double-ionization threshold. The slow transition towards chaos is analogous to the situation observed theoretically in doubly excited helium with  $^1S^e$  symmetry [19]. Second, the Rydberg series with a well-defined  $K$ -value were analyzed individually. In this way, the loss of quantum numbers  $N$  can be confirmed for Rydberg series with  $K \rightarrow K_{max}$  by the NNS distributions that clearly develop towards a Wigner distribution. The statistical NNS distribution below  $I_{17}$  matches almost a Wigner form, so that the transition from a Poisson-like distribution for a regular system towards a Wigner-like distribution for a chaotic system is almost completed. Since for Rydberg series with a well-defined  $K$ -value the angle between the two electrons and nucleus is constant, the 3-D helium atom can be considered to represent a stretched 1-D character with both electrons on opposite sites of the nucleus for  $K \rightarrow K_{max}$ . The Wigner-like form of  $K$ -selected NNS distributions for Rydberg series with  $K \rightarrow K_{max}$  shows a quantum signature of chaos for the stretched “1-D” cases of real 3-D helium. These “1-D” cases in 3-D helium agree very well with those obtained for 1-D helium as presented by Püttner *et al.* [18], and we confirm the prediction that a full Wigner distribution will be found around  $I_{17}$ . We also employed statistical tools for studies of the linewidths and the Fano  $q$  parameters, which exhibit a Porter-Thomas distribution and a Lorentzian form, respectively. Both distributions are considered to be characteristics of a chaotic systems, and the present statistical studies for the Fano  $q$  parameters are the first confirmation of the theoretical predictions [84]. In summary, a number of features typically for chaotic systems are found for the doubly excited states of helium. In order to answer the open question if Ericson fluctuations are present in the spectra of doubly excited helium, the cross section below a number of additional thresholds have to be studied experimentally and theoretically.

With time-of-flight electron spectrometer, the partial cross sections (PCS) below the SITs  $I_5$  to  $I_9$  [37, 104] and the angular distribution parameters (ADP) below the SITs  $I_5$  to  $I_7$  were performed. The measurements for the PCS below the SITs  $I_6$  to  $I_9$  were performed for the first time and show a very good agreement with eigenchannel R-matrix calculations [33]. It was shown that the PCSs display additional information about the doubly excited states of helium. For example, the resonance  $8, 4_{10}$  of the secondary series was resolved for the first time in the present PCS in contrast to the TCS, although the

latter one was measured with higher resolution and better signal-to-noise ratio. For the lower ionization thresholds we confirmed so-called general pattern in the PCSs which were explained theoretically by Schneider *et. al.* [33] with the propensity rules derived from a molecular description of helium. We also showed experimentally that these general patterns disappear with an increasing number of perturbers. In addition, an analytical expression for the mirroring behavior [36] was extended to a more general case. This allowed us to understand the mirroring behavior in doubly excited helium and to discuss it in a more general way: we realized that the mirroring behavior between two groups of PCSs can only occur accidentally, although  $\rho^2 \rightarrow 0$ . This is due to the fact that the amplitudes of the variations in the PCS and in the TCS are of the same order of magnitude. Further, the ADPs below the SITs  $I_5$  to  $I_7$  were measured, and the data below the SITs  $I_6$  to  $I_7$  are new. The additional resonance  $6, 2_8$  was observed in the present ADP measurements for the first time. All statistical studies for quantum chaos had to be done on the basis of theoretical results. The present experimental results for these PCSs and ADPs can however substantiate the applied theoretical methods, and, in this way, assure the validity of these statistical analysis on the basis of theoretical data. In addition, since the PCSs and the ADPs carry additional coupling informations of the outgoing channel, quantum chaos is expected to manifest itself more clearly or in different ways in these data as compared to the TCS. Vice versa, the success of the present experimental and theoretical study of TCSs in such a high excited region will initiate further studies of PCSs and ADPs.

In the last part of dissertation, detailed R-matrix calculations on the  $n$ - and  $l$ -specific PCSs and ADPs leading to all possible accessible ionization channels ( $nl$ ) were performed for the energy regions below the SITs  $I_3$  and  $I_4$ . In these regions, the approximate quantum numbers reflecting the strong electron correlation can be used to identify the spectra with physical meaning. The good agreement with the measurements proves the quality of the used target wave functions. In particular, it is encouraging to find almost perfect agreement between the present calculations [126] and very recent measurements [39, 40] of  $l$ -specific PCSs below the SIT  $I_4$ . Mirroring behavior for other series were also analyzed and similarity to the principal series were found with respect to mirroring behavior. As  $\rho^2 \rightarrow 1$ , we observe that the principal series below the SITs  $I_3$  and  $I_4$  mimic each other in the  $l$ -specific PCSs, which is also the reason why mimicking behavior is found for the principal series among the  $n$ - and  $l$ -specific ADPs.

In the near future, with the ball chamber which was used to measure the ADPs in this moment the no-dipole effect of doubly excited helium will be studied. The expected forward-backward asymmetry of the outgoing electrons relative to the propagation direction of light will allow to access the final states with a total symmetry different from  $^1P^o$ . In addition, the set-up of metastable helium source is on the way, which can be used for the observations of photoionization process of  $^3P^o$  doubly excited helium.