Introduction

The discovery of strong electron correlation effects in double-excitation resonant states of helium in an experiment by Madden and Codling in the 60's of the past century [1] triggered the development of group-theoretical and molecular adiabatic approximation approaches to understand this unexpected strong correlation dynamics described by a set of approximate quantum numbers in low doubly excited states. Close to the double-ionization threshold, the spectra exhibit complicated fluctuations due to electron correlation which finally leads to quantum signatures of chaos [2]. The aim of this dissertation is to study doubly excited helium in a very broad energy range from strong correlation dynamics below the lower ionization thresholds to quantum chaotic dynamics very close to the double-ionization threshold. In addition, the present studies are expected to improve the understanding of electron correlation, develop new concepts for quantum chaos, and also improve the understanding of quantum mechanics from the viewpoint of classical chaos.

Quantum chaos can be considered as the behavior of a quantum mechanical system whose classical counterpart would be chaotic. The field is not limited to theoretical interests related to the quantum-classical correspondence of chaos. Since the 1990's new techniques of fabricating semiconductor microstructures have opened a whole new active research area, namely the field of mesoscopic physics. A large number of interesting systems in the mesoscopic regime, e.g. quantum dots [3, 4] and tunneling devices [5], can be well understood on the basis of quantum chaos [6]. Gutzwiller's trace formula [7, 8] presents the correspondence between quantum mechanics and classical nonlinear mechanics of chaotic systems and has been successfully applied to a number of fields [9]. So far, most of the studies of chaos are limited to simple theoretical model problems with two degrees of freedom such as the double pendulum [10]. For atomic systems, intensive studies have been performed for the hydrogen atom in a strong magnetic field [11, 12, 13, 14, 15]. Very recently, the first experiment on the chaotic scattering of ^{85}Rb atoms was achieved [16, 17]. For doubly excited helium close to the double-ionization threshold, where $\hbar \to 0$, quantum chaos is expected since its classical counterpart, the classical three-body system, is a nonintegrable system. For this atom, theoretical studies of quantum chaos are mainly focused on 1-D helium [10, 18] and $^1S^e$ doubly excited helium [19], since these studies can be carried out with good accuracy for reasons of simplicity as compared to the $^1P^o$ doubly excited states in 3-D helium.

In this dissertation, the $^1P^o$ doubly excited states in helium are addressed. By absorbing a single photon, ground-state helium can be excited into a singly ionized continuum state $^1P^o$ by direct and indirect photoionization processes. As a consequence, Fano profiles [20] are observed for the resonances in the cross section due to interference between these two processes. For the studies from strong correlation to quantum chaos in doubly excited helium, two kinds of experiments were performed in the dissertation. One is the measurement of total cross sections (TCS) by monitoring the total ion yield with a gas cell [21]. This technique provides high count rates and allows to measure the spectra with small cross sections extremely close to the double-ionization threshold, where one expects quantum signatures of chaos. In contrast to the total ion yield measurements of the TCS, the measurements for partial cross sections (PCS) and angular distribution parameters (ADP) were performed by detecting photoelectron spectra with an advanced time-of-flight (TOF) technique [22]. This technique can distinguish between the decay channels to the different final states $He^+(n)$ by measuring the various kinetic energies of emitted electrons. For this topic, our studies focus on strong correlation dynamics of autoionization in doubly excited helium in the intermediate energy region, i.e. below the single ionization thresholds (SIT) I_5 to I_9. The SIT I_N implies the energy when the principal quantum number of the inner electron in doubly excited states is N and the one of the outer electron is infinite. Both kinds of measurements were performed using third-generation synchrotron radiation light sources. High brightness of the light and the high-resolution monochromators are prerequisites for performing these state-of-the-art experimental studies. Some fundamental knowledge about the atomic interaction with the electromagnetic field as well as about the properties of synchrotron radiation and monochromators is introduced in the first part of this dissertation.

Due to strong electron correlation inside the helium atom, the spectra of the double-excitation states cannot be described by traditional quantum numbers like e.g. the orbital angular momentum quantum number. Instead, a set of approximate quantum numbers were introduced for the description of the spectra [23]. It was later shown that these approximate quantum numbers are identical to the exact quantum numbers which describe the separable two-center Coulomb problem of H_2^+ [24]. In the energy region above I_4, the most interesting features are caused by perturbers; these are low lying resonances of Rydberg series belonging to higher ionization thresholds, which interfere with Rydberg series that converge towards a lower ionization threshold. Predicted by quantum defect theory [25], these perturbers will influence the energy positions of the Rydberg resonances by an increase of the quantum defect by one within the region of the interference; the linewidths and Fano parameters q of the corresponding Fano resonances are also modulated. In the region close to the double-ionization threshold, the interferences due to the overlap of several perturbers with different Rydberg series render the observed spectra highly complicated. In this case, the approximate quantum numbers reflecting strong electron correlation are expected to lose their physical meaning, and the regularities in the two-electron resonance spectrum start to dissolve. If many resonances in the spectrum are strongly overlapping and the spectra hence fluctuating, one expects the occurrence of Ericson fluctuations [26] in the spectra; they are, together with the loss of good quantum numbers, assumed to be features of quantum chaos. Close to the double-ionization threshold, the helium atom can be described in a semiclassical way. Considering that its classical counterpart is nonintegrable, one expects to see the manifestations of chaos in doubly excited helium from a study of quantum numbers, spectral fluctuations, and the statistical properties of resonance parameters. Therefore, the last eV below the double-ionization threshold has drawn considerable attention in both experimental and theoretical studies.

Recently, Püttner *et al.* have performed experimental and theoretical studies on the TCSs below the SIT I_9 with the result that the statistical properties of the energy levels revealed a transition towards quantum chaos [18]. In this dissertation, the experimental studies on the TCS are extended up to the SIT I_{15}, i.e. up to 6 further thresholds are measured. These new data will be analyzed together with the results of very recent state-of-the-art complex-rotation calculations of Delande [27] that were carried out up to the SIT I_{17}. This experimental and theoretical progress makes it possible to explore the exciting physical phenomenon—quantum chaos in doubly excited helium. On the basis of these data, the approximate quantum numbers, the question Ericson fluctuations as well as the statistical properties of energy levels, linewidths, and Fano parameters q will be topics in this dissertation. The statistical analysis displayed interesting precursor quantum signature of chaos in doubly excited helium. Many unexpected results were found and interesting questions such as the absence of Ericson fluctuations will be addressed. Semiclassical configurations [2, 28] related to the double-ionization dynamics in helium are employed to understand the chaotic dynamics of doubly excited resonances. Using the theoretical results, the experimental spectra will be assigned for the first time up to the SIT I_{14}. These highly interesting and novel results will be presented in Sect. 4 of the present dissertation. Up to now, the published experimental and theoretical studies of the TCS were limited to regions below the SIT I_9 [18, 29, 30, 31, 32, 33].

The studies of the PCSs and ADPs of doubly excited states of helium below the SITs I_5 to I_9 are performed with the TOF electron spectrometers and presented in Sect. 5 of this dissertation. These PCSs and ADPs show similarity which are called general pattern [34, 35]. They can be understood on the basis of propensity rules for autoionization decay [31, 33]. In this chapter, we will prove experimentally the presence of the general patterns up to the SIT I_9, but does not work well due to more and more perturbers. Interesting features, such as the mirroring behavior [36, 37, 38] of double-excitation resonances will also be addressed in this dissertation. Since the PCSs and the ADPs can carry additional information on the coupling of between outgoing channels, more resonances are expected to be observed in the PCSs than in the TCSs. Their clarifications can improve the understanding to the decay dynamics of doubly excited autoionization resonances. In addition, the PCSs and the ADPs provide additional tests of the quality of the theoretical data that are employed in our statistical study of quantum chaos [18].

Theoretical $n-$ and $l-$specific PCSs and ADPs as well as corresponding TCSs below the SIT I_4 were calculated by the R-matrix method; the results are given in the last part of dissertation. A justification of this theoretical work is the impressive agreement between the results of the present calculations and those of very recent measurements of the l-specific PCSs by Harries *et al.* [39, 40]. On the basis of reliable l-specific PCSs and ADPs, more general cross section patterns such as mirroring behavior and mimicking behavior [38] are expected to appear, which can further improve the understanding of correlation and decay dynamics in two-electron atoms.