New Interseries Interferences in Doubly-Excited Helium

R. Püttner[1], M. Domke[1], M. Martins[1], A.S. Schlachter[2], and G. Kaindl[1]

[1]Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
[2]Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA

INTRODUCTION

Doubly-excited helium has been a prototypical system for the study of electron-electron correlation since the pioneering work of Madden and Codling [1] and its interpretation by Cooper et al. [2] in 1963. In the early 1990's, the progress in synchrotron-radiation facilities and monochromator design led to a spectral resolution of ΔE=4.0 meV at the SX700/II monochromator at BESSY/Berlin [3,4], resulting in more detailed insight into the doubly-excited states of helium by resolving new Rydberg series as well as the first interseries interferences. Up to now the observation of 6 interseries interferences has been reported [5].

The resolution in the soft x-ray region was very recently improved to ΔE=1.0 meV at beamline 9.0.1 of the ALS/Berkeley. [6]. At the beginning, this improved resolution was used to reexamine the double excitations below the N=2 and N=3 ionization thresholds (N=quantum number of the inner electron). Here, we report on the observation of three hitherto unobserved interseries interferences below the N=4, 6, and 8 ionization thresholds.

EXPERIMENT

The experiments were performed at BL 9.0.1 of the ALS, using a 925-lines/mm spherical grating. The photoionization measurements were done with a gas cell containing two parallel charge-collecting plates of 10-cm active length, filled with He at pressures from 100 to 500 μbar, and separated from the monochromator vacuum by a 1200-Å thick carbon window. All spectra were normalized to the incident photon flux monitored with a gold grid in front of the gas cell. The spectra presented were recorded at a resolution of $\Delta E \cong 1.7$ meV.

RESULTS AND DISCUSSION

The optically excited $^1P^o$ double-excitation states of helium can be assigned by N,K_n, with N (n) denoting the quantum number of the inner (outer) electron and K the angular-correlation quantum number [7]. The 2N+1 Rydberg series converging towards the $He^+(N)$ ionization threshold can be distinguished by K=N-1, N-2, ..., -(N-1). Below most ionization thresholds, $He^+(N)$, only the most intense Rydberg series with K=N-2 (principal series) and the second most intense series (secondary series) are observed. Starting with N=5, the lowest Rydberg state is below the N-1 ionization threshold and can influence the N-1 Rydberg series as a perturber, leading to an interference. An interference is expected if N-K is equal for the two series, the Rydberg series and the perturber state [8]. We report on the three previously unresolved interferences of the principal series $4,2_n$ with the perturber state $5,3_5$ ($4,2_n/5,3_5$), the series $6,4_n$ with the perturber state $7,5_8$ ($6,4_n/7,5_8$), and $8,6_n$ with the perturber state $9,7_{11}$ ($8,6_n/9,7_{11}$).

Fig. 1. shows the interference of the Rydberg series $6,4_n$ with the perturber $7,5_8$. The solid line through the data points represents the fit results, with the solid (dotted) subspectrum describing the principal (secondary) series. The calculated energy position and natural linewidth of the perturber $7,5_8$ is also indicated [9]. The assignments of the Rydberg states are given by the bar diagrams on top of the spectrum. The spectrum in Fig. 1 show all characteristic features of an interference: due to the perturber, the intensities and the linewidths of the $6,4_n$ resonances exhibit a Fano-like behavior [5] decreasing from n=13-18 and subsequently increasing from

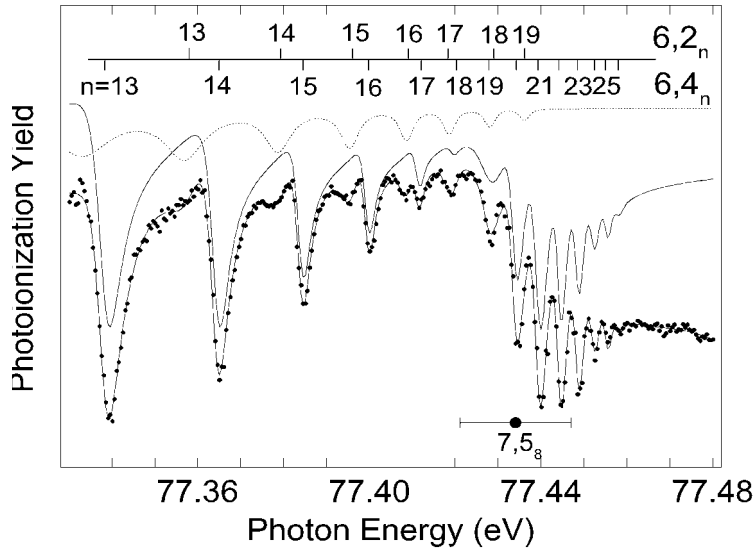

Fig. 1: The $6,4_n$ Rydberg series with the perturber $7,5_8$. The solid line through the data points represents the fit result. The solid (dotted) line represents the principal series $6,4_n$ (secondary series $6,2_n$). The theoretical position and natural linewidth of the perturber are also indicated. The assignment of the resonances is given in the vertical-bar diagram above the spectrum.

n=19-22. The reduced linewidth, defined as $\Gamma^*=\Gamma_n \times (n-\delta)^3$ (with Γ_n= natural linewidth and δ: quantum defect), is constant for an unperturbed Rydberg series, but it is influenced by the perturber leading to a Fano-like variation of Γ^* (q>1) [5]. Since one cannot distinguish between the perturber and the Rydberg series, the perturber is counted to the series, resulting in an increase of the quantum defect δ by 1 for the series. This can also be seen in the spectrum: between the $6,2_{17}$ and $6,2_{18}$ resonances of the unperturbed secondary series with constant quantum defect δ, two states of the principal series are observed, namely $6,4_{18}$ and $6,4_{19}$ resonances, which clearly demonstrates the described increase of the quantum defect by 1.

The principal series $4,2_n$ interacting with the perturber $5,3_5$ is shown in Fig. 2. The principal series $4,2_n$ is resolved up to n=23 and the secondary series $4,0_n$ up to n=17. The behaviors of the principal and secondary series are analogous to those of the series $6,4_n$ and $6,2_n$. However, this spectrum reveals a peculiarity: the perturber $5,3_5$ is calculated to be quite broad with Γ=59.2 meV and extends beyond the N=4 ionization threshold. Only the lower part of the interference is resolved, causing an increase of the quantum defect δ by less than one (see below).

Fig. 3 shows the $8,6_n$ principal series up to n=26 with the interferences with the $9,7_{10}$ and $9,7_{11}$ perturbers resolved. The interference $8,6_n/9,7_{10}$ has been studied previously [10], while the interference $8,6_n/9,7_{11}$ is observed in the present work for the first time.

The quantum defects δ derived from the energy positions of the $4,2_n$, $6,4_n$, and $8,6_n$ resonances are displayed in Fig. 4. The gray-colored area shows the energy region of the newly resolved interferences due to improved energy resolution. The quantum defects, calculated on the basis of quantum defect theory by using the theoretical energy positions and the natural widths of the perturber states [9], are indicated by the three solid lines. A good agreement between the observed and calculated quantum defects is clearly seen: As expected, the quantum defect increases by one in the energy regions of the interferences $6,4_n/7,5_8$ and $8,6_n/9,7_{11}$. In the energy region of the interference $4,2_n/5,3_5$, on the other hand, the quantum defect increases by only ≅0.5, due to the fact that the perturber is very broad and has also an effect above threshold.

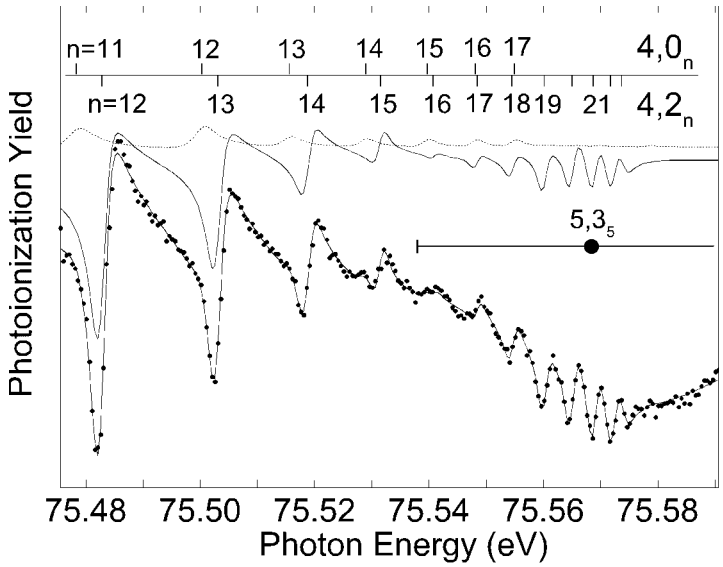

Fig. 2: The $4,2_n$ Rydberg series with the perturber $5,3_5$. The solid line through the data points represents the fit result. The solid (dotted) line represents the principal series $4,2_n$ (secondary series $4,0_n$). The theoretical position and natural linewidth of the perturber are also indicated. The assignment of the resonances is given in the vertical-bar diagram above the spectrum.

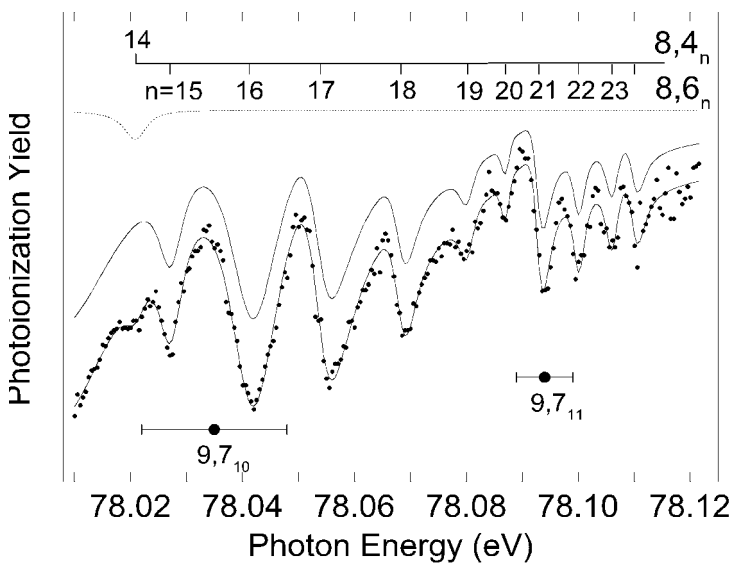

Fig. 3: The $8,6_n$ Rydberg series with the perturbers $9,7_{10}$ and $9,7_{11}$. The solid line through the data points represents the fit result. The solid (dotted) line represents the principal series $8,6_n$ (secondary series $8,4_n$). The theoretical positions and natural linewidths of the perturber are also indicated. The assignment of the resonances is given in the vertical-bar diagram above the spectrum.

As a consequence, the increase of the quantum defect is not limited to the energy range below threshold.

CONCLUSIONS

We studied three previously unresolved interseries interferences in helium. The derived quantum defects of the resonances behave as expected on the basis of quantum-defect theory.

ACKNOWLEDGEMENT

The authors appreciate valuable experimental assistance at BL 9.0.1 by J.D. Bozek.

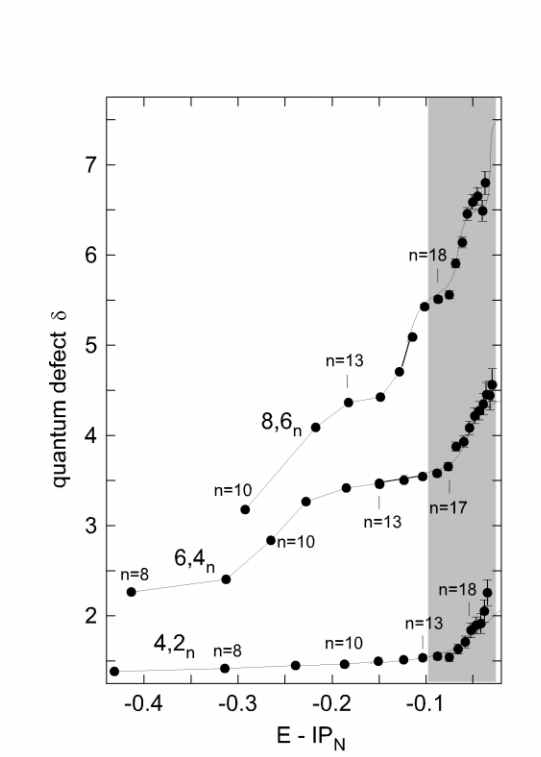

Fig. 4: Quantum defects δ of the resonances of the principal series $4,2_n$, $6,4_n$, and $8,6_n$ plotted as a function of the term energy, i.e. the energy difference to the relevant ionization threshold $He^+(N)$. The solid lines represent the calculated values of the quantum defect (see text).

REFERENCES

1. R.P. Madden and K. Codling, Phys. Rev. Lett. **10**, 516 (1963).
2. J.W. Cooper, U. Fano, and F. Prats, Phys. Rev. Lett. **10**, 518 (1963).
3. M. Domke, C. Xue, A. Puschmann, T. Mandel, E. Hudson, D.A. Shirley, G. Kaindl, C.H. Greene, H.R. Sadeghpour, and H. Petersen, Phys. Rev. Lett. **66**, 1306 (1991).
4. M. Domke, T. Mandel, A. Puschmann, C. Xue, D.A. Shirley, G. Kaindl, H. Petersen, and P. Kuske, Rev. Sci. Instrum. **63**, 80 (1992).
5. M. Domke, K. Schulz, G. Remmers, G. Kaindl, and D. Wintgen, Phys. Rev. A **53**, 1424 (1996).
6. G. Kaindl, K. Schulz, P.A. Heimann, J.D. Bozek, and A.S. Schlachter, Synchrotron Radiation News **8**, 29 (1995).
7. K. Schulz, G. Kaindl, M. Domke, J.D. Bozek, P.A. Heimann, A.S. Schlachter, and J.M. Rost, Phys. Rev. Lett. **77**, 3086 (1996).
8. A. Bürgers and D. Wintgen, J. Phys. B **27**, L131 (1994).
9. J. M. Rost, K. Schulz, M. Domke, and G. Kaindl, J. Phys B **30**, 4663 (1997).
10. M. Domke, K. Schulz, G. Remmers, A. Gutiérrez, G. Kaindl, and D. Wintgen, Phys. Rev. A **53**, R4309 (1995).

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Science, Materials Science Division, of the Department of Energy under Contract No. DE-AC03-76SF00098. The work in Berlin was supported by the Bundesminister für Bildung, Wissenschaft, Forschung und Technologie, project No. 05-650-KEA, and the Deutsche Forschungsgemeinschaft, project Do-561/1-1.

Principal investigator: G. Kaindl, Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin-Dahlem , Germany. E-mail: kaindl@physik.fu-berlin.de. Telephone: ++49-30-838-2977. Fax ++49-30-838-6560.