Evidence of an Excited Angular Correlation Mode in High-Lying He**

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(Received 18 June 1992)

We report an *ab initio* calculation of the photoionization spectrum of He in the energy regime hitherto unexplored theoretically, namely, energies near the $He^+(N)$ states up to $N = 7$. The recently reported experimental photoionization spectrum of He is fully reproduced by using our previously reported hyperspherical close-coupling method. We find clear evidence of an excited angular correlation mode that was presumably overlooked in the analysis of the experimental data. We also present a calculated spectrum below He^+ ($N = 7$) as a reference for future experiments.

PACS numbers: 32.70.Cs, 32.80.Fb

Helium is a prototype three-body system in atomic physics and plays an important role in the study of electron-electron correlations since correlation effects manifest themselves unhindered by the presence of core electrons. The photoionization spectra of He have long provided [1, 2] an important source of information on the electron-electron correlations. Despite numerous studies of energy levels, widths, and correlation patterns in the 1970s and 1980s, rigorous computational methods [3] aiming at high precision have been developed only recently to include the doubly excited resonances and the continuum contributions. However, calculating the photoionization spectra at photon energies above 75 eV is an extremely demanding task for the existing computational methods because of the presence of multitudes of resonances. On the experimental side, the recently published photoionization experiment [4] with synchrotron radiation has reportedly achieved energy resolution as high as 6 meV. Very rich resonance structures are revealed at high photon energies ($\sim$ 76 eV), including even interference between resonance series belonging to different N manifolds. Exploring these high-lying doubly excited states is important for consolidating our present understanding of the electron-electron correlations. This will in turn yield us vital information as to what takes place at even higher energies and how to describe double ionization near threshold. Theoretical reproduction of the experimental findings is a first step that we need to achieve for extracting relevant information and before we explore even higher energy regimes.

In this Letter, we report on the successful reproduction of the spectrum using the hyperspherical close-coupling (HSCC) method [5, 6]. Let us summarize the gist of the method. The hyperspherical coordinate method [7–9] has been used successfully to describe electron-electron correlation patterns in two-electron systems. What makes this method unique and powerful is the replacement of the independent particle radial distances r_1 and r_2 by a pair of collective variables. They are the hyperradius $R = \sqrt{r_1^2 + r_2^2}$ measuring the "size" of the electron pair and the hyperangle α=arctan $(\frac{r_2}{r_1})$ describing the degree of electron-electron radial correlation.

In our previous papers, we improved the adiabatic hyperspherical method into a dependable high-precision method. The method consists of the following six steps. (i) Set up a set of fast convergent and orthonormal diabatic channel functions in the α coordinate using node-adapted mesh points. (ii) Solve by the diabatic-by-sector method a set of close-coupling equations obtained by expanding over the diabatic channel functions. (iii) Match the solved wave functions to the asymptotic solutions by the two-dimensional matching procedure and determine the asymptotically correct K matrix. (iv) Impose the incoming wave boundary condition on the final-state wave function. (v) Evaluate the corresponding dipole transition matrix between initial and final states in both the length and acceleration forms. (vi) Calculate the electron-impact and photon-impact cross sections. The details will be published elsewhere shortly [5, 6].

We verified the accuracy and efficiency of the HSCC method for various processes [5, 6, 10] below the $N = 2$ threshold. Owing to the use of the fast and monotonically convergent diabatic basis functions, we have not encountered any practical difficulties that prevent us from studying the high-lying doubly excited states. Indeed, the 75-eV region analyzed by the experiment turned out to be well within the capacity of our method as we will show below. Even a higher-energy region seems manageable by the method. We will present one such example at the end of the paper. Let us now analyze details of the experimental spectrum for the high-lying doubly excited $^1P^o$ states of He.

We show in Fig. 1(a) the calculated photoionization spectrum. We included 49 diabatic basis functions and carried out the two-dimensional matching at $R = 170$ a.u. The length and acceleration forms gave results identical to three significant digits. The presented results are by the acceleration form. Detailed structures near threshold are shown on an expanded scale in Figs. 1(b) and 1(c).

The spectrum Gaussian convoluted with the resolution

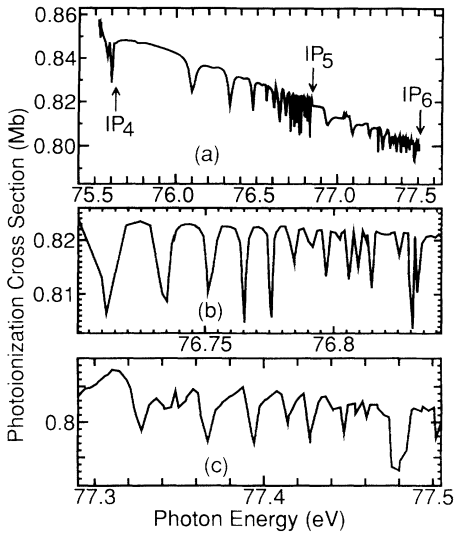

FIG. 1. Photoionization spectrum of He $^1P^o$ states: (a) below the $N = 5$ and 6 thresholds (IP_5, IP_6), (b) magnification of the high-n region below the $N = 5$ threshold, (c) magnification of the high-n region below the $N = 6$ threshold.

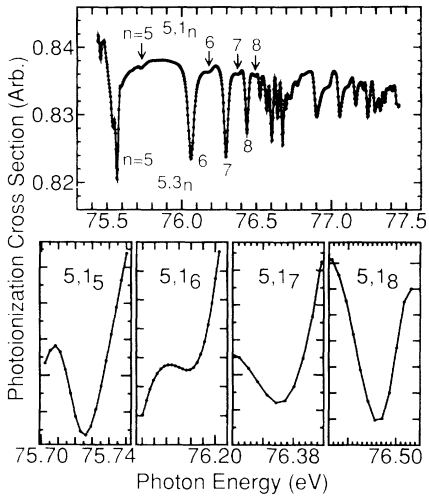

FIG. 2. Photoionization spectrum of He convoluted with experimental resolution of 6 meV below the $N = 5$ and 6 thresholds. Resonances labeled $5,1_n$ are magnified underneath. Note that the dots are calculated points and the solid lines simply join them smoothly.

of 6 meV is shown in Fig. 2 with the linearly decreasing background subtracted off. This background comes mainly from the $1s\epsilon p$ elastic partial cross section. The quantity subtracted at energy E is given by $aE + b$ with $a = 0.643$ Mb/a.u. and b=0.0672 Mb. The spectrum shown in this figure should be compared with the experimental one shown in Fig. 2(c) of Ref. [4]. Realize that every experimental detail identifiable is satisfactorily reproduced. Let us note that our result fits the experimental spectrum better than the multichannel quantum-defect theory fit does (Fig. 4 of Ref. [4]).

The salient features such as windows and dips are due to doubly excited resonance states. They provide us with information about the quasistability of the two-electron motion that leads to these long-lived states. It is now an established fact that the doubly excited states are classifiable using approximate correlation quantum numbers that permit to make a correspondence between the two-electron system and a floppy triatomic molecule. Typical modes of a triatomic molecule are the symmetric and antisymmetric stretching modes, and bending vibrational and rotational modes. The quantum numbers associated with each of these modes were analyzed earlier and are represented as $N(K,T)^A n$ [8, 11]. For the meaning of each symbol entering into this notation, the reader should consult Refs. [8, 11].

Following the experiments [4, 12], we use the simplified nomenclature N, K_n because all those resolved states have $T = 1$ and $A = +$. The dominant features seen in this energy range indeed correspond to autoionizing states belonging to the lowest $A = +$ type hyperspherical channel [4, 9]. However, the second dominant features that are indeed visible in the experimental spectrum, but overlooked in the analysis [4], belong to the second lowest $A = +$ type channel. These states actually pertain to excitation in the angular correlation mode that corresponds to the molecular bending vibrational mode. These resonances are identified as $5, 1_n$ states and displayed separately in Fig. 2.

Another interesting feature is caused by the overlap of two series belonging to different N manifolds. Such features are seen in Fig. 2 at photon energies between 76.5 and 76.8 eV and between 77.2 and 77.4 eV. In this energy range, the lowest doubly excited state in the N manifold manifests itself as an envelope of high-lying Rydberg series in the $(N - 1)$ manifold ($N > 4$). These may be better perceived as we use the effective principal quantum number instead of the photon energy. This is done in Fig. 3 with the former energy range as an example.

Near the $N = 4$ threshold, IP_4, we see the onset of the above-mentioned interference. The dominant feature is clearly identifiable as the $5,3_5$ state. However, the $6,4_6$ state appears as an envelope and at the same time perturbs the Rydberg series so strongly that there is an abrupt jump in the quantum defect at around n^*=7.5 [n^* is defined through $E = IP_N - (Z - 1)^2/2n^{*2}$]. In other words, we see here an abrupt increase in the density of states by one. So it may not be appropriate to assign to this sudden increase in the density of states the quantum numbers $5,3_{10}$ as done in Fig. 4 of Ref. [4]. In Fig. 3, we labeled this structure $6,4_6$ because it results from the interference between the $6,4_6$ state, and its energy coincides accidentally with that of the calculated $6,4_6$ state [13].

We calculated the positions and widths of the Wannier ridge states [i.e., $N, (N - 2)_N$ states] and fitted to

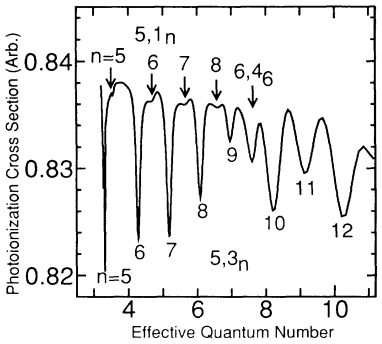

FIG. 3. Photoionization spectrum below the $N = 5$ threshold as a function of effective quantum number.

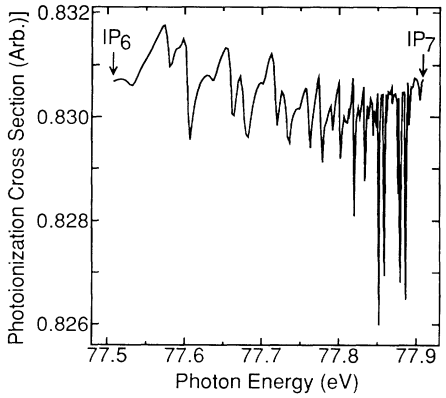

FIG. 4. Photoionization spectrum below the $N = 7$ threshold.

the two-electron Rydberg formula [14] $E(N,N) = -(Z - \sigma)^2/(N - \mu)^2$ for $N = 3,4,5,6$. The obtained values for the screening parameter σ=0.1592 and the quantum defect $\mu = -0.1774$ are very close to the experimental ones of σ=0.1587 and $\mu = -0.1815$. It is interesting to note that comparing the obtained screening parameter with that of H^- [9] indicates its independence from the atomic number Z. Should this indeed hold for Li^+ and for other He-like ions?

As a prediction and stimulation for future experimental works, we present also the spectrum below $N = 7$. The spectrum becomes very complex due to the strong interchannel interference. The result without convolution but with the background subtracted off is shown in Fig. 4. We see that there are at least three Rydberg series detectable. The correct assignment of quantum numbers needs more information than the total cross section. It remains a problem for further studies.

Once again, our HSCC method has proved very efficient and accurate. The electron-electron correlations are shown to be well described by the diabatic basis functions we proposed. The method can cover the energy range from the ground state up to these high-lying doubly excited states by merely augmenting the number of basis functions from 20 to 50; that is, no *ad hoc* readjustment is necessary. Note that we neglect the dipole attraction between the outermost electron and He^+ in the asymptotic matching functions. Although it is not important in the present case due to the predominance of the long-range Coulomb attraction, it may play a very important role for the spectrum of H^- whose analysis is currently in progress. Finally, our predicted cross section for the $N = 7$ manifold indicates that resonances due to higher excitation in angular correlation modes become visible. They are expected to overlap with resonances of higher-N manifolds. The spectrum would become exceedingly complicated. How will it evolve toward the double-ionization threshold? We appear to be seeing only the onset of a new regime of spectrum.

We acknowledge gratefully the support from the international cooperative research program between the National Science Foundation and the Japan Society for the Promotion of Science.

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