Decay Paths of Interfering Two-Electron Excitations in Helium

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Partial photoionization cross sections and photoelectron angular distributions of He in the region of interfering Rydberg series below the $n = 5$ threshold are measured and compared with theoretical results based on the hyperspherical close-coupling method. At a bandpass of 12 meV for the photon energy, this level of differentiation offers the most critical assessment of the dynamics of the two-electron excitations to date. A good understanding is achieved.

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Since their discovery by Madden and Codling [1] in the early 1960s, the double electron excitations into $^1P^0$ states in helium have served as the benchmark for the analyses, both experimental and theoretical, of the influence of electron correlation on the electron dynamics of this prototype three-body system. In recent years, great advances have been made experimentally [2–4], owing to the development of modern synchrotron radiation sources with their associated high-resolution monochromators, and theoretically [5–10], owing to the development of refined calculational methods. However, high-resolution work has remained restricted to total absorption measurements [2–4], and the desirable critical comparison with theory at the level of detail of partial photoionization cross sections and photoelectron angular distribution parameters has remained elusive. These properties can be delineated only by an *electron emission* measurement.

We report here the first electron emission study at a resolution approaching that of the less-revealing absorption measurements in the region where the interference of the higher members of the $n = 5$ Rydberg series with $n = 6$ excitations places the most exacting demands on both experiment and theory. We complement the data with a calculation of the pertinent partial cross sections and β parameters by an advanced *ab initio* method [7,10]. This comparison of theory with experiment provides the most detailed test to date of our understanding of the dynamics of the two-electron excitations in helium.

The experiment was performed on the undulator beam line 9.0.1 at the Advanced Light Source [11]. Our apparatus and procedures are described in detail elsewhere [12,13]. Briefly, electrons were detected simultaneously in two analyzers at $\theta = 0°$ and $90°$ with respect to the polarization direction and, additionally, at the pseudomagic angle $\theta_m = \frac{1}{2}\cos^{-1}(\frac{-1}{3P})$. The degree of linear polarization was determined to be $P = 0.97(1)$. At θ_m the *relative* partial cross sections $\sigma_n(\mathrm{rel})$ were obtained directly, and the data at $\theta = 0°$ and $90°$ yielded both the photoelectron angular distribution parameters β_n as well as $\sigma_n(\mathrm{rel})$. The acceptance half angle was 2.5(5)°. Spectra were recorded primarily in the constant ionic state mode (CIS) in which a $He^+(n)$ state is scanned synchronously with the photon energy. The resolution is then given by the photon bandpass, which was determined to be 12(3) meV by comparison with absorption data [2,4]. Spectra were normalized to the photon intensity and corrected for background and analyzer transmission. At a constant source pressure of 10^{-2} Pa, no pressure corrections were needed. The absolute photon energy was adjusted to the theoretical resonance positions [10], using $2\ \mathrm{Ry_{He}} = 27.207\,67$ eV and a double ionization potential of 79.003 44 eV. The experimental resonance positions of the corresponding absorption measurement [2,4] were reproduced with an overall shift of about 8 meV.

The theoretical approach is based on the hyperspherical close-coupling method described elsewhere [6] and was extended to provide differential cross sections and the β_n parameters in addition to the partial cross sections σ_n reported earlier [10]. Briefly, the hyperspherical coordinates describe the motion of the two electrons collectively in terms of (1) the hyperradius R, which represents the size of the two-electron system as a whole, (2) the hyperangle α, which represents the correlation between the radial motions of the two electrons, and (3) the four independent-electron angular coordinates. The total wave function of the initial (final) state of photoionization of helium was expanded in terms of 22 (55) basis functions generated from the wave equation in α and angular coordinates. When substituted into the Schrödinger equation for the total system, this wave function leads to close-coupling equations in R, which are referred to as hyperspherical close-coupling equations.

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They were solved outward to a large R (9 a.u. for the initial state and 200 a.u. for the final state), and the solution was matched to the asymptotic wave function in terms of the usual independent-electron coordinates. The energy of the initial state and the resonance energies converged to within a few meV. The partial cross sections calculated *ab initio* in this way without adjustable parameters are expected to be accurate to $\sim 1\%$ on the basis of the agreement of the length- and acceleration-gauge results within $\sim 1\%$.

The principal features of this photoelectron study are sketched in Fig. 1. A double excitation state of the $n = 5$ series decays into the $He^+(n)$, $n = 1$ to 4, states, and CIS scans for each ionic state yield the $\sigma_n(\mathrm{rel})$ on a relative basis and the β_n parameters, which are given by

$$\beta_n = \sum_l \sigma_{nl}\beta_{nl}/\sigma_n ,$$

where σ_{nl} and β_{nl} cannot be resolved. At a given photon energy, the $\sigma_n(\mathrm{rel})$ are given by a photoelectron spectrum (PES) as displayed in Fig. 1(a) for $h\nu$ just below the $n = 5$ threshold. The $\sigma_n(\mathrm{rel})$ values at $h\nu = 76.8$ eV are listed in Fig. 1(a) and represent an average between our narrow-bandpass data and the recent recommended values [14] based on broad-bandpass data. These relative values are converted to absolute partial cross sections $\sigma_n(\mathrm{abs})$ by normalization to $\sigma_{\mathrm{tot}} = 780$ kb at this energy as the recommended value [14], which utilizes the latest, most accurate measurements of σ_{tot}. This procedure allows us to make a sensitive comparison between experiment and theory for the absolute σ_n of each partial channel that can be populated by the decay of the $n = 5$ resonances.

The transition from the regular part of the $n = 5$ series into the region of interference with the first $n = 6$ resonance is displayed in Fig. 2 as it manifests itself in the $He^+(n = 2)$ channel. The resonances are identified in the $n(K,T)_i^A$ notation of the double excitations [16,17], where n refers to the ionization limit of the series, i is the running radial quantum number of the outer electron, and $(K,T)^A$ represent a set of correlation quantum numbers. The major $5(3,1)_i^+$ series and its perturbation by the $6(4,1)_6^+$ resonance are seen in the partial cross section $\sigma_{n=2}$, while the weaker series $5(1,1)_i^+$ is manifest in the partial $\beta_{n=2}$ parameter, and hitherto unobserved excitations, corresponding to the $5(4,0)_i^-$ series in which

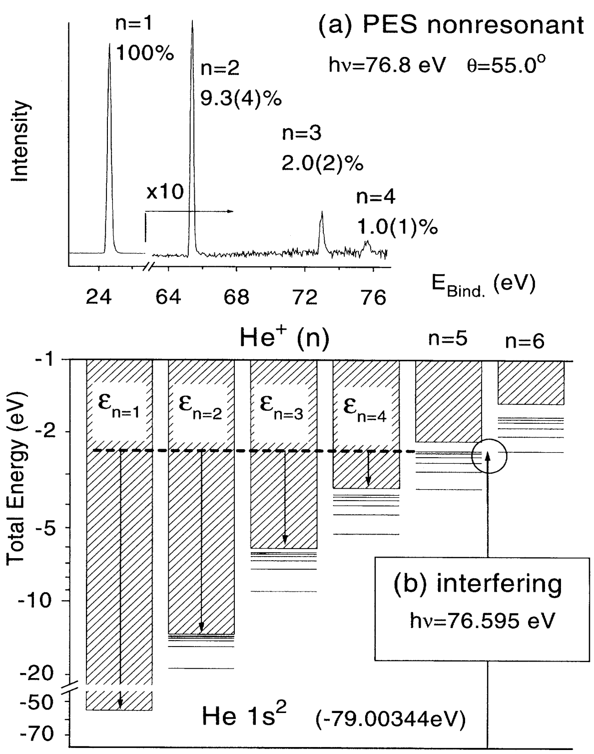

FIG. 1. (a) Nonresonant photoelectron spectrum of He recorded at the pseudomagic angle. The partial cross sections σ_n are given as a percentage of the main line. (b) Energy diagram of continua (shaded areas) and the first six discrete levels of the strongest double excitations. Note the overlap of the $n = 5$ series with the first $n = 6$ level. An excitation into the interference region is indicated by the arrow.

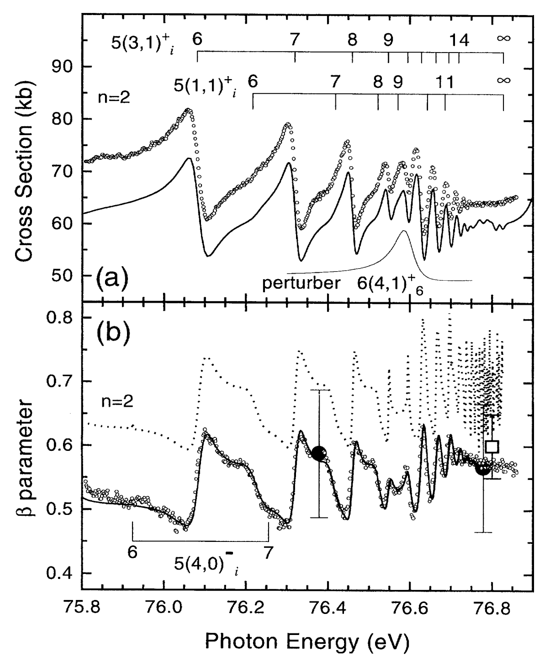

FIG. 2. The decay of the $5(K,T)$ series into the $He^+(n = 2)$ state. (a) Experimental (open circles) and convoluted theoretical (solid line) partial cross section $\sigma_{n=2}$. The shape and position of the perturber $6(4,1)_6^+$ inferred from the higher $n = 6$ series members $i = 7, 8$ (Ref. [10]) are indicated by a thin solid line. (b) Experimental (open circles) and theoretical (dotted line) $\beta_{n=2}$ parameters. The convoluted theoretical result (solid line) has been superimposed on the experimental data by shifting it by –0.12 units. Solid circles (this experiment) and the square (Ref. [15]) are β parameters derived from PES measurements.

one of the electrons changes its sense of rotation, may be discerned in the $\beta_{n=2}$ data. We note that the $5(1,1)_i^+$ resonances have otherwise been observed only in the latest high-resolution absorption spectra [4].

Figure 3 shows the experimental partial cross sections σ_n and the total cross section σ_{tot} along with the theoretical curves over the interference region. To highlight the excellent agreement in the resonance-induced variations we overlap the experimental and theoretical results by using different scales in some panels. The factors between the scales are 1.05 (total), 1.06 ($n = 1$), 0.915 ($n = 2$), and 1.0 ($n = 3, 4$), and lie generally within the uncertainties in the theoretical (see above) and experimental absolute cross sections. The experimental errors are evaluated to be 1.5% (total) and 2% ($n = 1$) according to [14], and 5% ($n = 2$) and 10% ($n = 3, 4$) according to the partition described above.

The accord between the experiment and the calculation is very good for the resonance-induced variations as well for the absolute values of σ_n. The largest variance is noted in $\sigma_{n=1}$. The accord is not as good for σ_{tot}, because the experimental σ_{tot} is the sum of the individual σ_n, which vary greatly in their resonance shapes and are subject to small displacements, up to about 2 meV, by the resettability limits from scan to scan. However, theory reproduces well the $\sigma_{\text{tot}}(\text{rel})$ obtained directly in an ion-yield measurement [2,4]. As seen from Fig. 3, the absolute variation of the partial cross sections σ_n induced by the resonance states is of a similar magnitude for all channels n, while the relative variation is the smallest for $n = 1$ and the largest for $n = 4$. Interestingly, the patterns exhibited in the regular series members, $i \leq 8$ (see also Fig. 2), are carried over into the interference regime, albeit in attenuated form due to the oscillatory character induced by the strong perturber $6(4,1)_6^+$.

Whereas the partial cross sections provide sensitive criteria for the understanding of the double-excitation dynamics, further insight is afforded by the β_n parameters presented in Fig. 4. The CIS recordings of the β parameters through the interference region are matched with our PES data near 76.8 and 76.4 eV. The error bars indicate the ranges by which the CIS scans might be shifted as *a whole*. Our narrow-band β_n values are in satisfactory agreement with available broad-bandpass data [19], except for one case [20]. The β_n are subject to possible shifts in energy, up to about 3 meV.

As is the case for σ_n, the accord between the theoretical and experimental results is good also for the β_n. This

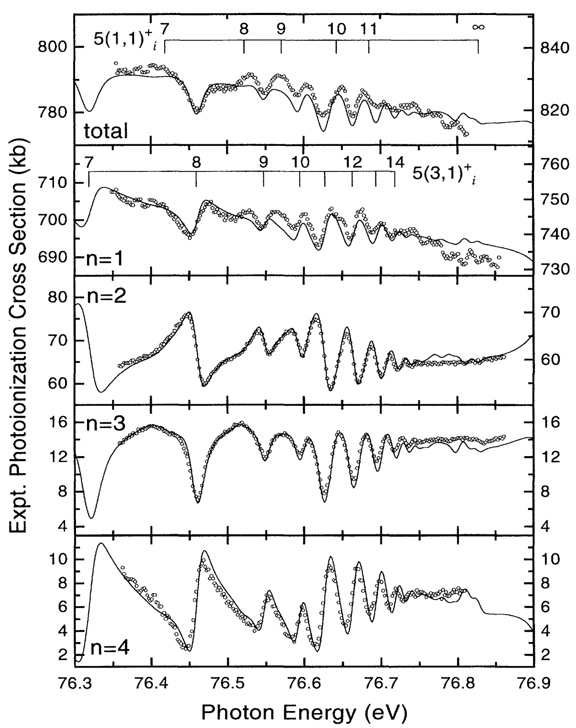

FIG. 3. Experimental (open circles) and convoluted theoretical (solid lines) total and partial absolute cross sections. The experimental data for $n = 1$ and the total cross section are smoothed. The left scale applies to the experimental data normalized to $\sigma_{\text{tot}} = 780$ kb at 76.8 eV (Ref. [14]), and the right scale applies to the theoretical results.

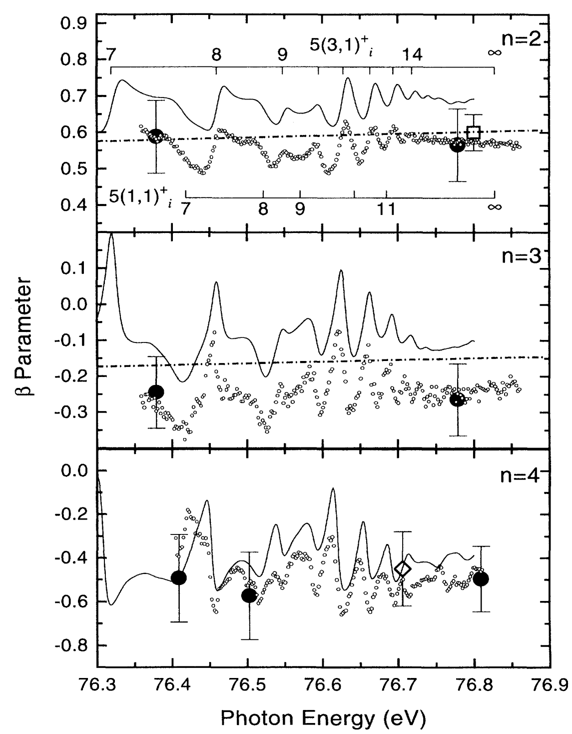

FIG. 4. Experimental (open circles) and convoluted theoretical (solid lines) β parameters. Experimental data for $n = 4$ are smoothed. Solid circles (this experiment), square (Ref. [15]), and diamond (Ref. [18]) represent β values derived from PES measurements. The dot-dashed lines are interpolated from data given in Ref. [19].

holds true for the amplitudes of the variations, the peak positions, and the resonance patterns. Differences are generally within the experimental uncertainties, except for the absolute magnitudes of the β_n, which slightly exceed these uncertainties for $n = 2$ and $n = 3$. Our results corroborate previous observations [18,19] and predictions [21] of β_n to decrease with increasing n and to approach -1 for high n and low kinetic energies due to the dynamically unfavored transitions [21].

In conclusion, we measured and calculated the partial photoionization cross sections and angular distributions of the four accessible final ionic states in helium in the photon energy regime of two interfering Rydberg series converging to $n = 5$ and $n = 6$. Overall, a very good agreement between experiment and theory was found, even on the scale of the absolute cross sections. The accord at this highly differentiated level demonstrates that we are now able to describe well the interactions and decays of the two-electron excitations in helium. Since the good accord extends to both the partial cross sections σ_n and the angular distribution parameters β_n, one might readily surmise that the unobservable σ_{nl} and β_{nl} are properly accounted for. Some questions merit further scrutiny. They pertain (1) to the finding that the resonance-induced variations in σ_n are of a similar magnitude for all n within the series and (2) to the resonance profiles which vary with n in a given series but display systematics [22] through the various $n(K,T)$ series. Comprehensive accounts of our work on the two-electron excitations in He will be presented [22,23] in the near future.

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