Very narrow doubly excited $2(1,0)^- n$ and $2(-1,0)^0 n\ ^1P^o$ states of helium

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We present a detailed theoretical study of the very narrow $A = -$ and $A = 0$ series seen in the recently reported high-resolution photoionization spectrum of He below the $N = 2$ threshold [M. Domke *et al.*, Phys. Rev. Lett. **69**, 1171 (1992)]. The hyperspherical close-coupling method combined with the multichannel quantum-defect theory is used and the photoionization cross section has been calculated over a wide energy range with high efficiency. Our results are in excellent agreement with the experimental data and compare favorably with recent accurate theoretical works.

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Since the first observation [1] of doubly excited states of He, many experimental [2] as well as theoretical [3–11] efforts have been focused on the photoionization spectrum of He. Although three Rydberg series of the $^1P^o$ symmetry ought to exist below the $N = 2$ threshold, only two of them, namely the $A = +$ and $A = -$ series have been experimentally observed while the third $A = 0$ series remained missing. The large disparity in intensity of the + and − series suggested a substantial difference in radial correlation patterns. The physical significance of this finding has long overshadowed the missing third series. The main experimental difficulty in locating the $A = 0$ series is its extreme narrowness. On the other hand, this narrowness has led to some confusion among theoretical works. While almost all the theoretical methods [6, 11] produced similar energy positions and widths for the $A = +$ and $A = -$ resonances, they produced quite different results for the $A = 0$ series [11, 12]. Some theoretical calculations predicted even an incorrect level-ordering of the $A = 0$ states with respect to the $A = -$ states [12].

Improvement in experimental resolving power has made it possible to observe the hitherto "missing" $A = 0$ states of He in a recent photoionization spectrum [12]. It is thus an important theoretical task to evaluate and analyze the quantum-defect parameters of this last series. The accuracy of various computational methods may be checked on this numerically stringent case. Previously, we proposed a general computational method [hyperspherical close-coupling (HSCC) method] for two-electron atoms [11]. The accuracy and efficiency of the method have been checked on various physical quantities of He [11, 13]. Also we have reproduced the experimental photoionization spectrum below $N = 5$ and 6 thresholds [14]. In this paper, we reproduce the experimental photoionization spectrum below $N = 2$ by combining our method with the multichannel quantum-defect theory (MQDT). In this article, we focus our attention mainly on the extremely narrow $A = 0$ series and check our method.

Our HSCC method is based on the use of the hyperspherical coordinates [15], which replaces the independent particle radial distances r_1 and r_2 by a pair of collective variables: R and α. The hyperradius $R = \sqrt{r_1^2 + r_2^2}$ measures the "size" of the electron pair and the hyperangle α=arctan$(\frac{r_2}{r_1})$ describes the degree of electron-electron radial correlation. The total wave function $\Psi(\mathbf{r}_1, \mathbf{r}_2)$ is expanded in terms of the diabatic basis functions $\{\phi_\mu(R; \alpha, \hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2)\}$ which we proposed in Ref. [11], namely

$$\Psi = (R^{5/2} \cos\alpha \sin\alpha)^{-1} \sum_\mu F_\mu(R) \phi_\mu(R; \alpha, \hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2). \tag{1}$$

This expansion is rapidly and monotonically convergent. The Schrödinger equation is then cast into close-coupling equations for $F_\mu(R)$, which are solved by a standard numerical integration method. The solutions are then transformed into the reactance matrix form, thereby the MQDT parameters are evaluated. The details of our method can be found in Refs. [11, 13].

We employ the MQDT treatment [16] near thresholds because the state density becomes very high and more importantly the resonances become very narrow. The treatment matches the close-coupling solutions to the standing wave asymptotic solutions, thus postponing the imposition of the asymptotic closed-channel boundary condition. This treatment has a major advantage because the reactance matrix K and the dipole matrix D_k depend weakly on energy. Interpolation of these slowly varying parameters permits efficient computation with sufficient accuracy. As an example, in Fig. 1(a) we show the four components of the D matrix in the length form near $N = 2$ threshold as well as the phase shifts, namely the arctangent of the eigenvalues of K matrix in Fig. 1(b). We see that indeed the D matrix and the phase shifts are smooth functions of energy (note the expanded scale in Fig. 1). Within the energy range from $E = -0.515$ a.u. to the $N = 2$ threshold, about 15 energy mesh points can guarantee enough accuracy for the calculation of photoionization cross sections.

For calculating the physical quantities, we need to impose the asymptotic boundary condition. The rectangular matrix L defined below eliminates the exponentially increasing Coulomb functions from the closed channels,

$$L = \begin{bmatrix} L_{oo} \\ L_{co} \end{bmatrix}$$

where the submatrices are

$$L_{oo} = I_{oo}, \tag{2}$$

$$L_{co} = (\tan \pi\nu_c + K_{cc})^{-1} K_{co} \tag{3}$$

where o and c designate sets of open and weakly closed channels, respectively, and the diagonal matrix ν_c denotes the absolute value of the reciprocal wave number of the continuum electron in the closed channels. The physical reactance matrix $\tilde{K}$ and dipole matrix $\tilde{D}$ are given by

$$\tilde{K} = K_{oo} - K_{oc} L_{co}, \tag{4}$$

$$\tilde{D} = D_o - D_c L_{co}. \tag{5}$$

After the above elimination of the weakly closed channels and imposing incoming-wave boundary condition on the final state, the photoionization cross section (in a.u.) for a transition from the initial state into one of the final open channels may be expressed as

$$\sigma_i = 8\pi^2 \alpha\omega \left| \sum_j \tilde{D}_j (1 - i\tilde{K})^{-1}_{ji} \right|^2 \tag{6}$$

where α is the fine-structure constant and ω is the photon energy.

In our calculation the ground-state wave function for He is also calculated by the HSCC method. With about 20 diabatic basis functions, the energy level obtained is $E_0 = -2.90358$ a.u. The calculation of the photoionization cross section is performed in both length and acceleration forms. The two forms give the same results within two digits with about 30 basis functions for the final $^1P^o$ state.

The doubly excited states may be classified by a set of quantum numbers $N(K,T)^A n$. Here K and T are the quantum numbers pertaining to the angular correlation [17] while A pertains to the radial correlation [18]. N is the principal quantum number of the inner electron and also represents the threshold of He^+ while n is the radial quantum number of the outer electron. In this scheme, the three Rydberg series of He ($^1P^o$) below $N = 2$ are denoted as $2(0,1)^+n$, $2(1,0)^-n$, and $2(-1,0)^0n$, respectively. In our discussions to follow, we use instead the simplified notation n, A to represent a doubly excited state.

The recent experiment [12] has achieved very high 4-meV energy resolution. With such high resolving power, the extremely narrow states of the $A = 0$ series as well as higher n states of the $A = +$ and $-$ series have been detected. Reproduction of the experimental spectrum for the $A = +$ series is not expected to be a difficult task for our method [11]. Indeed our calculated positions and shapes of doubly excited states are in excellent agreement with the experimental ones up to the highest resolved $n, +$ state. As an example, we display the calculated photoionization spectrum corresponding to the photon energy from 64.3 eV to 65.4 eV in Fig. 2(a) and also we compare it with the experimental result which is shown in Fig. 2(b). Our calculated spectrum has been convoluted with the experimental resolution of 4 meV. The experimental result appears to have a somewhat decreasing background. Nonetheless, the experimental spectrum

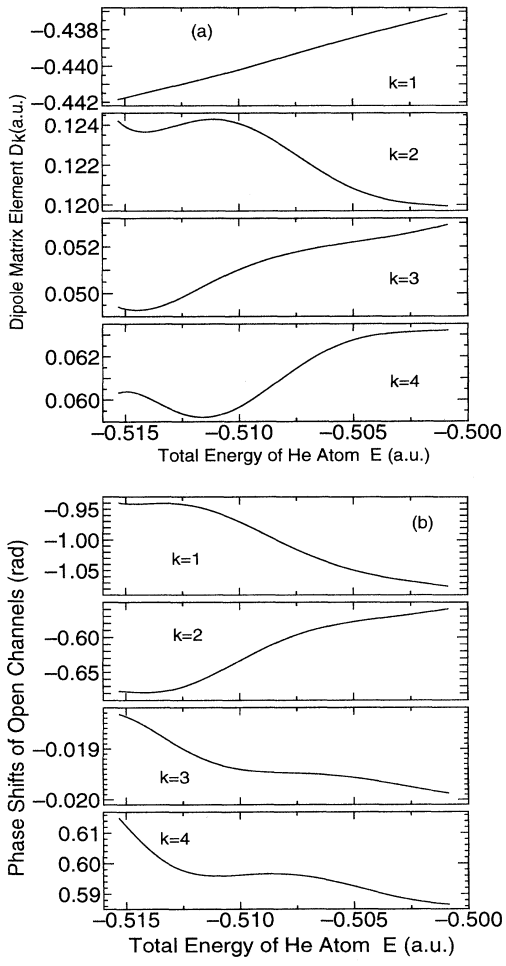

FIG. 1. Four components of (a) dipole matrix $D_k(E)$ in the length form and (b) phase shifts as a smooth function of total energy near the $N = 2$ threshold.

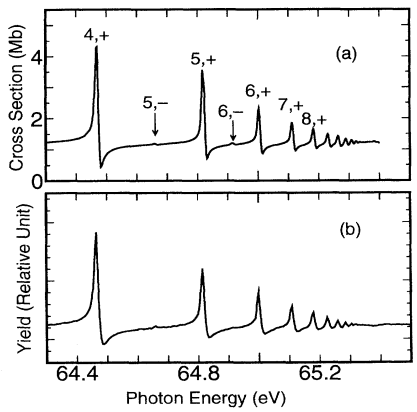

FIG. 2. Photoionization spectrum of He below the $N = 2$ threshold. (a) The present result and (b) the experimental one [12].

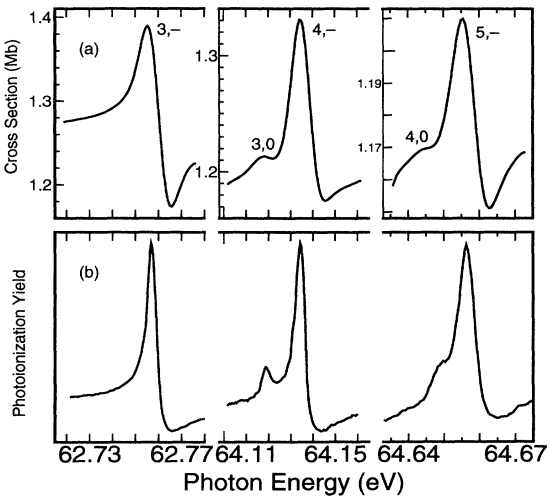

FIG. 3. The photoionization spectrum for $A = -$ and $A = 0$ resonance states. (a) The present results and (b) the experimental ones [12].

has been well reproduced.

The calculated spectrum is expanded for some of the very narrow $n,-$ and $n,0$ states as shown in Fig. 3(a) while the corresponding experimental result [12] is compared in Fig. 3(b). For the higher $n,0$ states, it becomes difficult to resolve them with 4-meV resolution power. Hence we give the calculated raw spectrum (that is the spectrum without any convolution) corresponding to the $7,-$ and $6,0$ states in Fig. 4 in order to indicate clearly the presence of the $6,0$ state. Here we wish to note that the intensity of the $A = 0$ series is of the same order of magnitude as that of the $A = -$ series. This can be also seen from Fig. 1 that the dipole moments corresponding to the transition from the ground state to these two series are of the same order of magnitude. The very reason why these $A = 0$ states have not been experimentally observed until recently [12] is not only a matter of photon intensity but due to the extreme narrowness which defied the finite experimental resolution.

In Table I, we compare our calculated widths of all the three series with those from other methods [6–10]. We see that the widths of the $A = +$ doubly excited states obtained by them are similar, while the results for the very narrow states, $A = 0$ and $-$, differ noticeably. Thus far, there has been no consensus as to which values are more accurate and as to which methods are more reliable. Because the widths of these very narrow states are

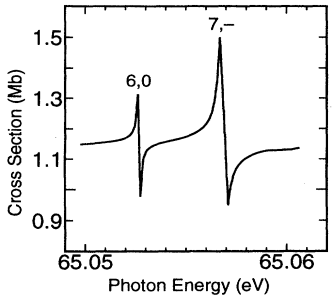

FIG. 4. The raw spectrum for $7,-$ and $6,0$ states.

100 times smaller than what the experiment can resolve, experiments with higher resolution power are still needed to check the accuracy of these theoretical values quantitatively. On the theoretical side, extracting the width by fitting to the spectrum becomes less and less reliable as the width itself becomes supernarrow. This may partly account for the disparity among the theoretical widths. However, as is clear from the comparison of our calculated spectrum with the experimental one in Fig. 3, we see that both the positions and shapes of these resonances are well reproduced. The accuracy and the capacity of our method are confirmed even in this difficult case.

Some benchmark results are presented for the resonance energies of $A = -$ and $A = 0$ states in the upper part of Table II. The error is estimated to be about 2 meV by varying the number of basis functions and the matching radius. Our results are in excellent agreement with both experimental ones [12] and other recently implemented accurate theoretical calculations [6–10]. The quantum defect of the $A = -$ series is calculated to be 0.730 at threshold and that of the $A = 0$ series is negative, -0.230, while the experimental results are 0.7205 and -0.260, respectively. It is found [12] that previous calculations [5] give quite different energy separations, ΔE, between the $n,-$ and $n-1,0$ states. What is surprising is that some of the results give positive values for ΔE while others give negative values. In the lower part of Table II, we compare our calculated ΔE values with the experimental ones as well as with those from other more recent theoretical methods. We see that all of these recently calculated results agree very well with the experimental one. We find, however, that the absolute positions of the very narrow states depend much more sensitively on the matching radius in our calculation than the energy separations do. The error in the

TABLE I. The widths Γ in eV for doubly excited $^1P^o$ states of He below $N = 2$ threshold. The number $a[b]$ denotes $a \times 10^b$.

			Γ			
State	Present	Ref. [6]	Ref. [7]	Ref. [8]	Ref. [9]	Ref. [10]
$2,+$	3.73 [−2]	3.78 [−2]	3.74 [−2]	4.02 [−2]	3.84 [−2]	3.70 [−2]
$3,+$	8.32 [−3]	8.27 [−3]	8.19 [−3]	8.92 [−3]	8.39 [3]	8.11 [−3]
$4,+$	3.48 [−3]	3.01 [−3]	3.51 [−3]	3.84 [−3]	3.58 [−3]	3.43 [−3]
$3,-$	1.16 [−4]	1.04 [−4]	1.05 [−4]	1.13 [−4]	1.12 [−4]	1.03 [−4]
$4,-$	5.21 [−5]	5.44 [−5]	5.58 [−5]	7.21 [−5]	5.68 [−5]	4.11 [−5]
$3,0$	4.41 [−6]	3.3 [−6]	4.4 [−7]	3.30 [−6]	1.56 [−6]	4.41 [−7]

TABLE II. Resonance energies of the $A = -$ and $A = 0$ states, and energy separations between $n, -$ and $n-1, 0$ states.

State(s)	Observed [12]	Present	Ref. [6]	Ref. [7]	Ref. [8]	Ref. [9]
			Resonance energy (eV)			
$3,-$	62.7580	62.758	62.7611	62.7611	62.761	62.757
$3,0$	64.1189	64.119	64.1217	64.1211	64.127	64.1211
$4,-$	64.1353	64.136	64.1377	64.1374	64.142	64.134
$4,0$	64.6485	64.647	64.6514	64.6512	64.654	64.648
$5,-$	64.6574	64.657	64.6598	64.6598	64.662	64.656
$5,0$	64.9071	64.907		64.9096	64.820	64.910
$6,-$	64.9123	64.913		64.9145		
$6,0$	65.051	65.052		65.0545	65.003	
$7,-$	65.0552	65.056		65.0578		
$7,0$		65.143			65.112	
$8,-$	65.1435	65.146				
			Energy separation ΔE (meV)			
$4,-/3,0$	16.4(4)	17	16.0	16.3	16.0	16.0
$5,-/4,0$	8.9(6)	10	8.4	8.6	8.0	8.0
$6,-/5,0$	5.2(7)	6		4.9		
$7,-/6,0$	4(2)	4		3.3		
$8,-/7,0$		3				

energy separation is estimated to be about 1 meV. This error is smaller than that in energy position due to cancellation of errors. This is not difficult to understand because the separations are very small and the convergence of the expansion in Eq. (1) is monotonic. We find further that the convolution affects the peak position of each resonance by as much as 1 meV. Since the experimental positions are extracted from the convoluted cross sections, this suggests that the agreement with the experiment may be actually even better than presented in Table II.

To conclude, the photoionization spectrum of He below $N = 2$ threshold has been studied by combining our HSCC method and the MQDT. The experimental findings are well reproduced. The positions and shapes of the narrow $A = -$ series and even the extremely narrow $A = 0$ series agree with the experimental results. With the observation of the third series and with its successful reproduction at the 1-meV order, we have a firm understanding of the He($N = 2$) manifold.

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