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A NEW INVESTIGATION OF THE $^1S_e$-AUTOIONIZING STATES OF He AND H

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A NEW INVESTIGATION OF THE $^1S_e$-AUTOIONIZING STATES OF He AND H$^-$

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ABSTRACT

The present investigation should provide some insight into the convergence properties of some particular upper roots of the full Hamiltonian. In this connection detailed calculations using the scaling-variation orthogonalization procedure with different subsets of the basis up to order 54 have been carried through both for He and H$^-$. Subsets including basic functions up to f-g are used, and a complete mapping of the ten lowest roots of the energy matrix by a continuous variation of the scale factor has been carried out within a definite range. The investigation clearly demonstrates the stabilizing ability of particular upper roots throughout the extension of the basis if the orthogonalization is properly achieved. These roots in the second-step solution may be associated with certain autoionizing states (quasi doubly-excited, discrete states). The eigenvalues, obtained by optimizing the scale factor for these roots, are compared with those obtained for resonance positions in He$^+$ and H elastic-scattering calculations by using other approximate methods, and by experiments.

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I. INTRODUCTION

Much work has been done both theoretically and experimentally in connection with the determination of the positions of autoionizing states. The dual nature of such states, due to the autoionization process itself resulting in autoionized states on one hand, and due to the inverse process of electron capture resulting in compound states (resonance states) on the other hand, has been recognized for a long time since the pioneer works in the 1930's -- theoretical works by Fano\textsuperscript{1} and experimental by Whiddington, Priestly and Beutler.\textsuperscript{2} Many approximate methods have recently been devised\textsuperscript{3} for calculating numerically the energies of the autoionizing states; but, as expected, none has yielded the same accuracy and reliability that the bound-state variation method did when applied to the stationary states.\textsuperscript{4} However, when experimental values of the positions of the two lowest autoionizing states of the specific symmetry $1s^e$ of He became known,\textsuperscript{5} we were a little puzzled about the relatively wide discrepancy between the theoretical value obtained by the truncated scaling-variation orthogonalization procedure\textsuperscript{6} for the lowest state and the experimental value, whereas we noticed a striking agreement for the next state. This fact stimulated us to make a rediagonalization of the 1958 matrix of order 20. Unfortunately, because of the far too slow and small computer used at that time, the lowest roots were not mapped for the twentieth-order matrix as was done both for the fourth-order and the tenth-order matrices.

We have now completely mapped all the ten lowest roots of the same twentieth-order matrix by a continuous variation of the scale parameter within a certain range. This re-examination showed that the results denoted with asterisks in Table I of the 1958 investigation\textsuperscript{6} are erroneous both for He and H\textsuperscript{−}. A corrigendum of these errors has been submitted for publication in Proc. Phys. Soc. (London).
The erroneous results of the (2s2s) roots have obviously led to some misleading conclusions in the literature about the ability of the truncated orthogonalization procedure to ensure bounds on the true energies of autoionizing states. It has often been argued that the convergence of the procedure will break down when more states in the basis are taken into account. The primary purpose of this new investigation is to demonstrate that there exist bounds to some upper roots of the energy matrix when the basis is enlarged and an effective orthogonalization is achieved. In view of the apparent stabilization property of these particular roots, they will be interpreted as representing the autoionizing states.
II. THE TRUNCATED ORTHOGONALIZATION PROCEDURE

The scaling-variation orthogonalization procedure with explicit formulae for the matrix elements is completely outlined in previous papers.\(^6\) The basis used in this new investigation is the same, namely a truncated expansion in Legendre polynomials

\[
\psi(x_1, x_2) = \sum_{q=0,1,j\geq q+1}^{M} c_{q(i,j)} \left\{ \varphi_i(x_1) \varphi_j(x_2) + \varphi_j(x_1) \varphi_i(x_2) \right\} P_q(\cos \theta_{12})
\]

where the \(\varphi\)'s are taken from the entirely discrete and complete set of the associated Laguerre functions of order \((2q + 2)\) with a single orbital exponent. These basic functions form a complete set if only the exponential parameter of the radial orbitals of either electron is the same throughout the basis.\(^7\) In addition, the set is entirely discrete, a fact which makes a small continuum component of the hydrogenic states present in nearly all the basic functions.\(^8\) Notice that the amount of this component depends on the scaling. The procedure itself is thus also devoted to a configuration interaction with the continuum.

In this investigation we have used the same exponential parameters for both electrons throughout the basis. This restriction on the trial functions makes good approximations for the \((1s1s)\)-, \((2s2s)\)- and \((2p2p)\)-states, but not so good for states associated with different electron configurations of the same symmetry.

Unfortunately, no rigorous variational method exists for the autoionizing states (quasi-bound stationary states) as it does for normalizable bound stationary states. The reason is that these states are not eigenstates of the full time-independent Hamiltonian, but, merely, that these states are represented by time-dependent solutions which induce radiationless transitions...
from the initial, doubly-excited, discrete states to the adjacent ionized states with equal energies. However, the energy of such an autoionizing state must be conserved during the very short period before decay, the lifetime for autoionizing states of order $10^{-14} - 10^{-12}$ sec.; i.e., the eigenvector representing the state must be considered as time-independent during this period. In view of this fact, application of a certain variational procedure to these states may be justified.

As is well known, when using a variational procedure in which all the parameters, both nonlinear and linear, occurring in the trial functions are optimized, the trial functions of the state of interest must be made orthogonal to all the lower states of the full Hamiltonian. In addition, the trial functions must satisfy certain boundary conditions, which means that for an autoionizing state they to some extent must satisfy asymptotically the outgoing boundary conditions with nonvanishing electron current at infinity. This will be the case when components of the hydrogenic continuum states are mixed into the trial functions, as automatically will be done when using our basis.

Furthermore, diagonalization of the energy matrix with respect to the truncated M-order basis is equivalent to optimizing the linear parameters by the variational method, thus obtaining M different roots, which, according to the Hylleraas-Undheim theorem, are upper bounds to the M lowest states of the system under consideration. This fact is still true when nonlinear parameters are being optimized.

In quantum mechanics it is also asserted that roots belonging to states of equal symmetry (equal L, S, and parity) cannot intersect, whereas roots belonging to states of different symmetry can intersect. This statement holds in any case in which the Hamiltonian contains a parameter to be varied. For the
autoionizing states with an infinity of continuum states of equal and lower energy and an infinite number of lower-lying bound discrete states of identical symmetry, the truncated scaling-variation procedure is no longer justified on the basis of the variational theory. However, the results of this investigation show that by using subsets containing, in addition to \( p-p, d-d, f-f \) and \( g-g \) states, a sufficient number of \( lsn s \) states, where \( n \geq 1 \) relative to the number of \( nsn's' \) states, where \( n \neq n' \geq 2 \) in a ratio greater than 1.0, for example, the roots representing the \((2s2s)^-\) and \((2p2p)^-\) configurations start out from the beginning even before roots belonging to the infinite number of higher configurations appear in the picture. These roots come out already with a fourth-order subset including the basic functions \( 1s1s, 1s2s, 2s2s \) and \( 2p2p \). These roots also seem to be stabilized by the extension of the subsets in the basis, provided the above mentioned ratio is approximately satisfied. This unusual turnup of upper roots (second-step roots) is probably achieved by the equal scaling of both electrons, and therefore also by an approximate orthogonalization to lower states. The results of this investigation clearly demonstrate that these roots will approach certain limits when the basis is enlarged. However, we cannot consider these limits as upper bounds.

The optimization of the scale parameter (the common single orbital exponent) does not destroy the stabilization (orthogonalization) as often has been argued in connection with this orthogonalization procedure. The question remains as to whether and to what extent the stabilization tendency of the upper roots will change if two orbital exponents, one for each electron, are optimized separately by extension of the basis. The basis itself will still be complete, and the particular roots will probably not change appreciably, but many more first-step roots will appear.
III. RESULTS AND DISCUSSION

As mentioned above, the purpose of this investigation is to give some insight into the convergence property of the truncated scaling-variation orthogonalization procedure. This has been done by diagonalization of the full Hamiltonian matrix with respect to a number of different subsets of the basis, and a continuous scaling variation of all the ten lowest roots has been performed within the range 0.20 to 0.50 of the parameter. The Hamiltonian contains this scaling parameter, and the eigenvalues are consequently functions of that parameter. The roots of the states of identical symmetry cannot intersect, and this fact is clearly demonstrated throughout the investigation with curves (roots) moving nicely apart in the range used.

The unit of length \( \frac{1}{2} a_0 Z^{-1} \) and the unit of energy \( Z^2 \text{Ry} \) have been used throughout the investigation. We shall consider in detail only the results of the He calculations because those of \( \text{H}^- \) turned out to possess quite the same peculiarities.

The numerical evaluation of the matrix elements has been performed in double-length, floating-point arithmetic; whereas, when diagonalizing the energy matrices, single-length arithmetic proved to be sufficient.

First of all we have demonstrated the convergence property of the second-step roots without restricting the states included in the basis. We have chosen the states in subsequent order: 2p2p to 2p6p, 3p3p to 3p6p, 4p4p to 4p6p, 5p5p, 5p6p, 3d3d to 3d6d, 4d4d to 4d6d, 5d5d, 5d6d, 4f4f to 4f6f, 5f5f, 5f6f, 5g5g, 5g6g (30), 2s2s (31), 2s3s to 2s6s (35), 3s3s to 3s6s, 4s4s to 4s6s, 5s5s, 5s6s (44), 1s1s (45), 1s2s (46), 1s3s, 1s4s (48), 1s5s to 1s10s (54).
In the list given above the numbers in the brackets designate the orders of the truncated subsets used, and the results obtained are illustrated in Figs. 1, 2 and 3. The numbers given in the brackets associated with the curves, however, designate the root numbers.

At this point it should be remembered that all the basic states are mutually orthogonal and that the interaction integral \( \langle \psi_1 | H | \psi_j \rangle \) is very large between the \( s-s \) states mutually compared to that occurring between \( s-s \) states and \( p-p, d-d, \) etc. states.

Some interesting aspects emerge from the results given in the figures. As can be seen, the behavior of the roots belonging to subsets up to order 35 is quite reasonable. These sets include the states \( 2s2s, 2s3s, 2s4s, 2s5s \) and \( 2s6s \) in addition to all the functions of the angular basis. All the roots seem to have only one minimum within the range. Probably several minima have moved up a little from the thirtieth-order subset. The interaction integrals are small for nearly all these states. If we now enlarge the subset to order 44 by the inclusion of more \( nsn' \) states, a strange behavior of the curves occurs. The behavior of several of the higher roots turns out like a two-step one, and some roots seem to have two marked minima. Some repulsion points appear between the higher roots. It is tempting to make the curves cross here, but according to the mentioned statement this is impossible. By now we have no explanation which is quite satisfactory for this strange behavior, but it looks as if it is caused by the inclusion of too many higher \( s-s \) states, where \( n \geq 2, n' \geq 4 \) and \( n \neq n' \). It is reasonable to assume that the inclusion of higher basic states, which are not orthogonal to the lower states, will cause a breakdown of the higher roots by variation of the scale parameter. We know that the higher \( s-s \) states overlap significantly the lower hydrogenic discrete
and continuum states, and this overlapping depends strongly on the scaling. One explanation might be that this interaction causes the roots to represent different states at different scaling. For instance, the specific root 44 (5) represents an upper state at $\eta = 0.38$ and a much lower state at higher scaling, i.e., for smaller values of $\eta (< 0.20)$. It looks as if this strange behavior is due to the total lack of orthogonality to lower states.

If we now gradually include the lower states $l$ns$n$ where $n \geq 1$ in the wave function, i.e., if we let the orthogonalization work, we notice that certain roots move up a little; and the upper step of, for example, root 46 (6) shows a marked minimum at $-0.3785 Z^2\text{Rh}$, whereas root 46 (7) indicates an upper step with no definite minimum at about $-0.31 Z^2\text{Rh}$.

Proceeding further now by including the $1s3s$ and $1s4s$ states in the basis, we get the subset of order 48, and the results are shown by the solid curves in Fig. 4. Some of the repulsion points are marked by dashed lines, and it should be noted that here the curves are really crossing in the case of the unperturbed problem, but no intersection will take place when the perturbation is taken into account. An appreciable change of the roots, probably associated with the two lowest autoionizing states, is apparent, namely 46 (6) or 46 (5), 46 (7) and 48 (6) and 48 (7). This change indicates a marked minimum for the upper step of 48 (7), whereas that of 48 (6) appears more distinct. In the forty-eighth-order subset only four $l$ns$n$ states are included compared to a number of fourteen doubly excited states of type $n\text{s}s'$, $n' \neq n$.

Figure 5 shows the results obtained by using the fifty-fourth-order subset where six additional $l$ns$n$ states have been included in the basis. As can be seen, the two-step behavior of the upper roots in the varied range has nearly disappeared, and the stabilized roots in this energy region seem to be the roots 9, 10 and probably 11.
It should be emphasized that an effective orthogonalization to lower states is achieved when a sufficient number of l sns states are included in the basis (in the fifty-fourth-order subset the ratio between lower and higher nan's states, n ≠ n', is 1.0), and it is plausible that this orthogonalization stabilizes the second-step roots.

We have investigated many subsets in which the above-mentioned ratio is smaller than 1.0. For comparison we give the results of a tenth-order subset in Fig. 6. As can be seen the same two-step behavior appears. If we exclude the states 2s4s, 2s5s and 2p4p and replace these by the states 3s3s, 3p3p and 3d3d, the normal behavior again occurs. The twentieth-order subset from 1958 with the ratio equal to 1.3, the results of which are demonstrated in Fig. 7, clearly shows normal behavior of the roots, and the stabilized roots seem to be numbers 5, 6 and 7.
IV. CONCLUSIONS

The conclusions drawn on the basis of this extended investigation can be formulated as follows:

(a) Restriction must be imposed on the ratio \(N(1sns)/N(nsn's)\) where \(N(1sns)\) is the number of singly-excited states and \(N(nsn's)\) is the number of higher doubly-excited states in the basis where \(n \neq n'\). It seems that this ratio must be larger than one if an effective orthogonalization to the lower states can be expected. This condition is fulfilled using the submatrices \((4 \times 4), (10 \times 10)\) given in Ref. (6) and \((20 \times 20)\) and \((54 \times 54)\). The stabilized roots associated with the two lowest autoionizing states are apparently \((3,4), (4,5), (5,6)\) and \((9,10)\), respectively. In Table I we have listed the values of these roots for optimum values of the scale parameter.

A similar extensive investigation has been performed for \(^{1}\text{H}^{-}\). This investigation showed exactly the same behavior of the roots throughout the enlargement of the basis. We only give the results of the \((20 \times 20)\) and \((54 \times 54)\) submatrices plotted as curves in Figs. 8 and 9, and in Table I the corresponding energies are listed for \(^{1}\text{H}^{-}\) as for \(^{6}\text{He}\). The most interesting result here is that, finally, with the fifty-fourth-order subset, the root probably associated with the second autoionizing state has a minimum which lies below the second threshold of hydrogen; i.e., the state is bound. In the same table a detailed comparison is made with results obtained by other approximate methods.

We have made a comparison for the ground state (the lowest root in the first step) in Table II, and as can be seen our fifty-fourth-order subset already gives very good agreement with the exact value
(b) It seems that optimization of the scale parameter does not destroy the orthogonalization if restriction (a) is imposed on the truncated basis. In all instances, the investigation clearly shows that the second-step roots converge to certain limits.

(c) Because the eigenvalues obtained by this truncated scaling-variation method are not necessarily upper bounds to the true energies of autoionizing states, the question remains of how these bounds converge when the entire complete basis is included. Recently, this question has come under discussion\(^9\) as to whether this procedure in the limit will approach the autoionizing states or the ionized states of the two-electron systems. According to the results obtained for the coefficients of the eigenvectors for different subsets when an effective orthogonalization is achieved, it looks as if the coefficients decrease appreciably for the higher doubly-excited states in the basis, even for the s-s states. The coefficients of the ground state, however, decrease even more. Therefore, we assume that the total component of the hydrogenic continuum will be small compared to that of the doubly-excited hydrogenic states, and thus it is reasonable that the procedure in the limit will approach the autoionizing states.

(d) The investigation shows that the variational procedure can be used with success if a reasonable choice of basic states has been made. It is important to note that as long as an eigenstate is orthogonal to the infinity of continuum states with equal and lower energy and to the infinite number of lower-lying discrete states, the corresponding eigenvalue is an upper bound of the true energy.
An exact orthogonalization process may be performed in principle if the entire basis has been included, and in this case the basis actually defines two different projection operators P and Q. One of the operators, say P, will because of the exact orthogonality, project out of the total wave function all the components originating in the lower discrete states and in the ionized states up to and including the adjacent state which is accessible through decay from the autoionizing state. On the other hand, the projector Q retains in a subspace all the states of double excitation of the independent approximation model, as well as the ionized states with higher energy. Hence, there will exist an interaction with the background continuum (more correctly with a part of it -- let us say a little window between the projected states in the Feshbach formalism) which is necessary to satisfy asymptotically the outgoing boundary conditions of the eigenstate.

In view of all this, it is possible that the energy shift due to the coupling with the continuum, to some extent, is included in the eigenvalues obtained by the truncated scaling-variation orthogonalization procedure; but the magnitude of this important quantity can only be estimated if we for the fifty-fourth-order subset in the basis, project out all the components originating in the hydrogenic ground-state configurations and then perform a similar variational procedure. In this way we should be able to estimate the shift caused by the upper background continuum. If we now interpret the minima of the roots 5 and 6 in Fig. 6 and of the roots 6 and 7 in Fig. 4 as if they really cross each other at the repulsion points, we obtain the results listed in Table III.
The interaction with the upper part of the continuum comes mostly into play by the additional basic states $1\text{s}_n\text{s}$, $n \geq 5$, in the fifty-fourth-order subset. A comparison of the results given in Tables I and III indicates that the energy shift for the $2s^2$ state seems to be larger; i.e., the width of this state should be larger, a fact which agrees with the close-coupling results.
ACKNOWLEDGMENTS

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The calculations involved in the investigation have been carried out on a UNIVAC 1107, located at the Norwegian Computing Center, Norway.
REFERENCES


   269 (1963).


TABLE CAPTIONS

Table I - A: Eigenvalue Minima (Second-Step Roots) in $Z^2$ Ry, and Interpretation.

B: Comparison of Available Data.

The positions are given in eV above ground state.


c) The most accurate position of the resonance state (W. McGowan, private communication).


e) Recalculated by including the 3d state.


Table II - Comparison of eigenvalues obtained for the ground state (fifty-fourth-order subset). Units are $Z^2$ Ry.


Table III - Eigenvalue Minima (second-step roots) of partly orthogonalized subsets. Units are $Z^2$ Ry.
### TABLE I

<table>
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TABLE II

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TABLE III

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FIGURE CAPTIONS

Figure 1 - Energy versus the scale parameter (subsets from 30 to 46).
          Units Rh = Ry.

Figure 2 - Energy versus the scale parameter (subsets 35 and 44).
          Units Rh = Ry.

Figure 3 - Energy versus the scale parameter (subsets 35, 44 and 46).
          Units Rh = Ry.

Figure 4 - Energy versus the scale parameter (forty-eighth-order subset).
          Units Rh = Ry.

Figure 5 - Energy versus the scale parameter (fifty-fourth-order subset).
          Units Rh = Ry.

Figure 6 - Energy versus the scale parameter (tenth-order subset).
          Units Rh = Ry. The states are: 2p2p, 2p3p, 2p4p, 2s2s, 2s3s,
          2s4s, 2s5s, 1s1s, 1s2s, 1s3s.

Figure 7 - Energy versus the scale parameter (twentieth-order subset, 1958).
          Units Rh = Ry.

Figure 8 - Energy versus the scale parameter (twentieth-order subset, 1958,
          Z = 1). Units Rh = Ry.

Figure 9 - Energy versus the scale parameter (fifty-fourth-order subset, Z = 1).
          Units Rh = Ry.
Figure 4
Figure 6
Figure 9