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Generalized Sturmian Solutions for Many-Particle Schrödinger Equations[†]

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The generalized Sturmian method for obtaining solutions to the many-particle Schrödinger equation is reviewed. The method makes use of basis functions that are solutions of an approximate Schrödinger equation with a weighted zeroth-order potential. The weighting factors are especially chosen so that all the basis functions are isoenergetic with the state they are used to represent. This means that all the basis functions have turning points located in such a position that they can contribute usefully to the synthesis of the wave function. The method is illustrated by a simple example—the calculation of atomic spectra.

Introduction

The Schrödinger equation for an N-particle system can be written in the form

$$\left[-\frac{1}{2}\Delta + V(\mathbf{x}) - E_\kappa\right]\Psi_\kappa(\mathbf{x}) = 0 \tag{1}$$

where

$$\mathbf{x} = \{\mathbf{x}_1, \mathbf{x}_2, ..., \mathbf{x}_N\} \tag{2}$$

$$\mathbf{x}_j \equiv \{x_j, y_j, z_j\} \qquad j = 1, 2, ..., N \tag{3}$$

and

$$\Delta \equiv \sum_{j=1}^{N}\frac{1}{m_j}\nabla_j^2 \tag{4}$$

It is usual to build up the solutions to (1) in terms of a set of basis functions, $\Phi_\nu(\mathbf{x})$:

$$\Psi_\kappa(\mathbf{x}) = \sum_\nu \Phi_\nu(\mathbf{x})C_{\nu,\kappa} \tag{5}$$

In the generalized Sturmian method,[1−26] these basis functions are chosen to be solutions to an approximate N-particle Schrödinger equation of the form

$$\left[-\frac{1}{2}\Delta + \beta_\nu V_0(\mathbf{x}) - E_\kappa\right]\Phi_\nu(\mathbf{x}) = 0 \tag{6}$$

Here V_0 resembles V as closely as possible (remembering, of course, that one must be able to solve (6)), and the constants β_ν are weighting factors especially chosen in such a way as to make the energy eigenvalue E_κ in (6) equal to the energy of the desired solution to (1). The advantage of this type of basis set is that in the asymptotic limit where both V and V_0 become small, all of the basis functions obey the same Schrödinger equation as the desired solution, since in this region

$$\left[-\frac{1}{2}\Delta + V(\mathbf{x}) - E_\kappa\right]\Psi_\kappa(\mathbf{x}) = 0 \rightarrow \left[-\frac{1}{2}\Delta - E_\kappa\right]\Psi_\kappa(\mathbf{x}) = 0 \tag{7}$$

and

$$\left[-\frac{1}{2}\Delta + \beta_\nu V_0(\mathbf{x}) - E_\kappa\right]\Phi_\nu(\mathbf{x}) = 0 \rightarrow \left[-\frac{1}{2}\Delta - E_\kappa\right]\Phi_\nu(\mathbf{x}) = 0 \tag{8}$$

Thus all the basis functions have the right asymptotic behavior, and their turning points occur in the right place, allowing them to contribute usefully to the synthesis of Ψ_κ. By contrast, conventional methods often represent Ψ_κ by superpositions of basis functions corresponding to various energies, and these may have the wrong behavior in the asymptotic region,

$$\left[-\frac{1}{2}\Delta + V_0(\mathbf{x}) - E_\nu\right]\Phi_\nu(\mathbf{x}) = 0 \rightarrow \left[-\frac{1}{2}\Delta - E_\nu\right]\Phi_\nu(\mathbf{x}) = 0 \tag{9}$$

as well as inappropriate turning points.

Application to the Calculation of Atomic Spectra

The generalized Sturmian method can be applied to a wide variety of problems in atomic and molecular physics. We can illustrate it with a particularly simple example—calculation of the spectra of few-electron atoms, in the nonrelativistic approximation, neglecting the motion of the nucleus as well as spin−orbit coupling. This case is especially simple because if V_0 is chosen to be the Coulomb attraction of the nucleus

$$V_0(\mathbf{x}) = -\sum_{j=1}^{N}\frac{Z}{r_j} \tag{10}$$

then (6) can be solved exactly[1] by Slater determinants of the form

$$\Phi_\nu = |\chi_{n,l,m,m_s}\chi_{n',l',m',m_s'}\chi_{n'',l'',m'',m_s''}...| \tag{11}$$

In eq 11, the one-electron functions χ_{n,l,m,m_s} are hydrogen-like

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spin–orbitals corresponding to an effective charge Q_ν, which is related to E_κ and to the principal quantum numbers in the configuration by

$$Q_\nu = \beta_\nu Z = \left(\frac{-2E_\kappa}{\frac{1}{n^2} + \frac{1}{n'^2} + \dots}\right)^{1/2} \tag{12}$$

To see that (11) will then be an exact solution of (6), we notice that each of the hydrogenlike spin–orbitals in (11) obeys a one-electron Schrödinger equation of the form

$$\left[-\frac{1}{2}\nabla_j^2 + \frac{Q_\nu^{\ 2}}{2n^2} - \frac{Q_\nu}{r_j}\right]\chi_{n,l,m,m_s}(\mathbf{x}_j) = 0 \tag{13}$$

Thus the kinetic energy operator acting on (11) yields

$$\begin{aligned}\left[-\frac{1}{2}\Delta\right]\Phi_\nu(\mathbf{x}) &= \\ &\left[-\frac{Q_\nu^{\ 2}}{2}\left(\frac{1}{n^2} + \frac{1}{n'^2} + \dots\right) + \frac{Q_\nu}{r_1} + \frac{Q_\nu}{r_2} + \dots\right]\Phi_\nu(\mathbf{x}) \\ &= [E_\kappa - \beta_\nu V_0(\mathbf{x})]\Phi_\nu(\mathbf{x}) \end{aligned} \tag{14}$$

Comparing this with eq 6, we can see that if Φ_ν has the form indicated by (11)–(13), it will be an exact solution of (6). One can show[16] that whenever $\beta_\nu \neq \beta_{\nu'}$ the configurations Φ_ν and $\Phi_{\nu'}$ will then obey a potential-weighted orthonormality relation of the form

$$\int \mathrm{d}x\ \Phi^*_{\nu'}(\mathbf{x})\ V_0(\mathbf{x})\ \Phi_\nu(\mathbf{x}) = \delta_{\nu',\nu}\frac{2E_\kappa}{\beta_\nu} \tag{15}$$

When two different configurations have the same value of β, a basis set transformation sometimes needs to be made to ensure that (15) holds.

Generalized Sturmian Secular Equations

It is convenient to introduce the variables

$$p_\kappa \equiv \sqrt{-2E_\kappa} \tag{16}$$

and

$$\mathscr{R}_\nu \equiv \sqrt{\frac{1}{n^2} + \frac{1}{n'^2} + \dots} \tag{17}$$

In terms of them, (12) can be rewritten in the form

$$Q_\nu = \beta_\nu Z = \frac{p_\kappa}{\mathscr{R}_\nu} \tag{18}$$

while the energy becomes

$$E_\kappa = -\frac{p_\kappa^{\ 2}}{2} \tag{19}$$

The potential V that enters the N-electron Schrödinger eq 1 includes both the nuclear attraction term V_0 and an interelectron repulsion term V':

$$V(\mathbf{x}) = V_0(\mathbf{x}) + V'(\mathbf{x}) = -\sum_{j=1}^{N}\frac{Z}{r_j} + \sum_{i>i}^{N}\sum_{i=1}^{N}\frac{1}{r_{ij}} \tag{20}$$

TABLE 1: ^{1}S Excited State Energies (in Hartrees) for the 2-Electron Isoelectronic Series[a]

	He	Li$^+$	Be^{2+}	B^{3+}	C^{4+}	N^{5+}
1s2s ^{1}S	−2.1429	−5.0329	−9.1730	−14.564	−21.206	−29.098
basis2	−2.1442	−5.0348	−9.1759	−14.567	−21.209	−29.102
exp	−2.1458	−5.0410	−9.1860	−14.582	−21.230	−29.131
1s3s ^{1}S	−2.0603	−4.7297	−8.5099	−13.402	−19.406	−26.521
basis2	−2.0606	−4.7301	−8.5107	−13.403	−19.407	−26.522
exp	−2.0611	−4.7339	−8.5183	−13.415	−19.425	−26.548
1s4s ^{1}S	−2.0332	−4.6276	−8.2837	−13.003	−18.785	−25.629
basis2	−2.0333	−4.6277	−8.2840	−13.003	−18.785	−25.630
exp	−2.0334	−4.6299	−8.2891			−25.654
1s5s ^{1}S	−2.0210	−4.5811	−8.1806	−12.820	−18.500	−25.220
exp	−2.0210	−4.5825				−25.241
1s6s ^{1}S	−2.0144	−4.5562	−8.1250	−12.721	−18.346	−24.998
exp	−2.0144	−4.5571				
1s7s ^{1}S	−2.0105	−4.5412	−8.0917	−12.662	−18.253	−24.865
exp	−2.0104	−4.5418				
1s8s ^{1}S	−2.0080	−4.5315	−8.0701	−12.624	−18.194	−24.779
exp	−2.0079					
1s9s ^{1}S	−2.0063	−4.5248	−8.0554	−12.598	−18.153	−24.720
exp	−2.0062					
1s10s ^{1}S	−2.0051	−4.5201	−8.0449	−12.579	−18.124	−24.678
exp	−2.0050					
1s11s ^{1}S	−2.0042	−4.5166	−8.0371	−12.566	−18.102	−24.647
exp	−2.0041					
1s12s ^{1}S	−2.0034	−4.5140	−8.0312	−12.555	−18.086	−24.624
exp	−2.0034					

[a] In making Table 1, the basis set used consisted of 63 generalized Sturmians. Singlet and triplet states were calculated simultaneously, 0.5 S of 499 MHz Intel Pentium III time being required for the calculation of 154 states. Basis 2 consisted of 245 symmetry-adapted configurations[20] including angular correlation up to $l_{\max} = 4$. Having constructed the interelectron repulsion matrix, eq 26, one can use the same matrix for any value of nuclear charge. Experimental values are taken from the NIST tables.[27]

TABLE 2: Doubly Excited Singlet S States of the 2-Electron Isoelectronic Series[a]

	C^{4+}	N^{5+}	O^{6+}	F^{7+}	Ne^{8+}	Na^{9+}
2s2s ^{1}S	−8.2861	−11.4133	−15.0406	−19.1678	−23.7949	−28.9221
2p2p ^{1}S	−7.6460	−10.6520	−14.1580	−18.1639	−22.6698	−27.6757
exp	−7.6531	−10.6604	−14.1699	−18.1777	−22.6908	−27.7075
2s3s ^{1}S	−6.0563	−8.3257	−10.9563	−13.9479	−17.3007	−21.0145
exp	−6.0618	−8.3317	−10.9642	−13.9618	−17.3220	−21.0470
2p3p ^{1}S	−5.8049	−8.0200	−10.5961	−13.5333	−16.8315	−20.4907
exp	−5.8105	−8.0252	−10.6018	−13.5441	−16.8478	−20.5122
2s4s ^{1}S	−5.3537	−7.3373	−9.6334	−12.2420	−15.1631	−18.3967
2p4p ^{1}S	−5.2453	−7.2056	−9.4786	−12.0641	−14.9622	−18.1729
2s5s ^{1}S	−5.0370	−6.8897	−9.0324	−11.4651	−14.1878	−17.2006
2p5p ^{1}S	−4.9810	−6.8216	−8.9522	−11.3730	−14.0838	−17.0846

[a] These are autoionizing states embedded in a continuum. The few available experimental energies[28−30] agree well with the calculated energies (in hartrees) shown in the table. Vainshtein and Safronova[29,30] give wavelengths for transitions to states that are listed in the NIST tables,[27] and from these the experimental energies were deduced. Solution of the Sturmian secular equations with basis 2 (Table 1) yielded the energies of all the ^{1}S states shown in Tables 1−5.

Substituting the expansion of the wave function (5) into (1), we obtain

$$\sum_\nu\left[-\frac{1}{2}\Delta + V(\mathbf{x}) - E_\kappa\right]\Phi_\nu(\mathbf{x})C_{\nu,\kappa} = 0 \tag{21}$$

We now make use of the fact that each of the basis configurations Φ_ν obeys eq 6. This allows us to rewrite (21) in the form

$$\sum_\nu[V(\mathbf{x}) - \beta_\nu V_0(\mathbf{x})]\Phi_\nu(\mathbf{x})C_{\nu,\kappa} = 0 \tag{22}$$

Multiplying (22) on the left by a conjugate configuration from

TABLE 3

	C^{4+}	N^{5+}	O^{6+}	F^{7+}	Ne^{8+}	Na^{9+}
3s3s ^{1}S	−3.7116	−5.1069	−6.7244	−8.5641	−10.6261	−12.9102
3p3p ^{1}S	−3.5578	−4.9241	−6.5126	−8.3231	−10.3559	−12.6108
3d3d ^{1}S	−3.2860	−4.5981	−6.1323	−7.8887	−9.8674	−12.0683
3s4s ^{1}S	−2.9098	−4.0011	−5.2661	−6.7046	−8.3168	−10.1025
3p4p ^{1}S	−2.8134	−3.8850	−5.1302	−6.5489	−8.1411	−9.9070
3d4d ^{1}S	−2.6940	−3.7379	−4.9554	−6.3463	−7.9107	−9.6488
3s5s ^{1}S	−2.5687	−3.5248	−4.6319	−5.8902	−7.2996	−8.8601
3p5p ^{1}S	−2.5165	−3.4620	−4.5586	−5.8062	−7.2050	−8.7549
3d5d ^{1}S	−2.4563	−3.3885	−4.4719	−5.7064	−7.0921	−8.6288

TABLE 4

	C^{4+}	N^{5+}	O^{6+}	F^{7+}	Ne^{8+}	Na^{9+}
4s4s ^{1}S	−1.7189	−2.3635	−3.1107	−3.9603	−4.9125	−5.9671
4p4p ^{1}S	−1.6754	−2.3116	−3.0503	−3.8914	−4.8350	−5.8811
4d4d ^{1}S	−1.6169	−2.2405	−2.9665	−3.7949	−4.7258	−5.7592
4f4f ^{1}S	−1.5517	−2.1588	−2.8682	−3.6801	−4.5943	−5.6111
4s5s ^{1}S	−1.5293	−2.0997	−2.7604	−3.5113	−4.3525	−5.2840
4p5p ^{1}S	−1.5031	−2.0683	−2.7238	−3.4695	−4.3055	−5.2318
4d5d ^{1}S	−1.4682	−2.0263	−2.6746	−3.4132	−4.2420	−5.1611
4f5f ^{1}S	−1.4308	−1.9801	−2.6198	−3.3497	−4.1700	−5.0806
4s6s ^{1}S	−1.4170	−1.9429	−2.5517	−3.2434	−4.0180	−4.8755
4p6p ^{1}S	−1.4004	−1.9230	−2.5285	−3.2169	−3.9881	−4.8423
4d6d ^{1}S	−1.3790	−1.8974	−2.4986	−3.1827	−3.9496	−4.7994
4f6f ^{1}S	−1.3553	−1.8684	−2.4646	−3.1436	−3.9056	−4.7504

TABLE 5

	C^{4+}	N^{5+}	O^{6+}	F^{7+}	Ne^{8+}	Na^{9+}
5s5s ^{1}S	−1.0065	−1.3957	−1.8448	−2.3470	−2.9096	−3.5327
5p5p ^{1}S	−0.9995	−1.3851	−1.8241	−2.3234	−2.8832	−3.5033
5d5d ^{1}S	−0.9858	−1.3606	−1.7957	−2.2911	−2.8470	−3.4632
5f5f ^{1}S	−0.9577	−1.3268	−1.7564	−2.2466	−2.7972	−3.4083
5g5g ^{1}S	−0.9479	−1.3003	−1.7196	−2.2039	−2.7486	−3.3537
5s6s ^{1}S	−0.9387	−1.2959	−1.7084	−2.1721	−2.6914	−3.2663
5p6p ^{1}S	−0.9336	−1.2894	−1.6958	−2.1578	−2.6754	−3.2486
5d6d ^{1}S	−0.9327	−1.2834	−1.6887	−2.1497	−2.6662	−3.2382
5f6f ^{1}S	−0.9261	−1.2746	−1.6787	−2.1385	−2.6538	−3.2247
5g6g ^{1}S	−0.9186	−1.2653	−1.6676	−2.1256	−2.6391	−3.2082
5s7s ^{1}S	−0.9056	−1.2504	−1.6510	−2.1073	−2.6192	−3.1867
5p7p ^{1}S	−0.8989	−1.2408	−1.6385	−2.0919	−2.6010	−3.1657
5d7d ^{1}S	−0.8967	−1.2296	−1.6263	−2.0786	−2.5865	−3.1501
5f7f ^{1}S	−0.8888	−1.2292	−1.6140	−2.0512	−2.5407	−3.0944
5g7g ^{1}S	−0.8883	−1.2195	−1.6029	−2.0391	−2.5387	−3.0825

TABLE 6: Energies for Doubly Excited States of C^{4+} with Varying Angular Correlation[a]

C^{4+}	$l_{\max} = 0$	$l_{\max} = 1$	$l_{\max} = 2$	$l_{\max} = 3$	$l_{\max} = 4$
2s2s ^{1}S	−8.1172	−8.2854	−8.2860	−8.2861	−8.2861
2p2p ^{1}S	−5.9810	−7.6058	−7.6400	−7.6449	−7.6460
2s3s ^{1}S	−5.3211	−6.0557	−6.0562	−6.0563	−6.0563
2p3p ^{1}S	−5.0202	−5.7949	−5.8038	−5.8048	−5.8049
2s4s ^{1}S	−4.8586	−5.3534	−5.3537	−5.3537	−5.3537
2p4p ^{1}S	−4.7620	−5.2411	−5.2448	−5.2452	−5.2453
2s5s ^{1}S	−4.6997	−5.0369	−5.0370	−5.0370	−5.0370
2p5p ^{1}S	−4.6572	−4.9789	−4.9808	−4.9810	−4.9810

[a] As can be seen from Table 1, the inclusion of angular correlation has only a small effect on the singly excited states. However, Table 6 shows that angular correlation is crucial for accurate determination of the energies of doubly excited states.

the basis set, and integrating over space and spin coordinates, we obtain

$$\sum_{\nu} \int dx\ \Phi^*_{\nu'}(\mathbf{x})[V(\mathbf{x}) - \beta_\nu V_0(\mathbf{x})]\Phi_\nu(\mathbf{x}) C_{\nu,\kappa} = 0 \qquad (23)$$

We now introduce the definition

$$T^0_{\nu',\nu} \equiv -\frac{1}{p_\kappa}\int dx\ \Phi^*_{\nu'}(\mathbf{x})\ V_0(\mathbf{x})\ \Phi_\nu(\mathbf{x}) \qquad (24)$$

With the help of the potential-weighted orthonormality relation (15) and making use of (18), we have

$$T^0_{\nu',\nu} = \delta_{\nu'\nu} Z \mathscr{R}_\nu \qquad (25)$$

Thus we see that the matrix $T^0_{\nu',\nu}$ defined by (24) is independent of p_κ. Similarly, it can be shown[16] that

$$T'_{\nu',\nu} \equiv -\frac{1}{p_\kappa}\int dx\ \Phi^*_{\nu'}(\mathbf{x})\ V'(\mathbf{x})\ \Phi_\nu(\mathbf{x}) \qquad (26)$$

is also independent of p_κ. Substituting these definitions into (23) and dividing by p_κ, we obtain the generalized Sturmian secular equations

$$\sum_{\nu}[\delta_{\nu',\nu} Z\mathscr{R}_\nu + T'_{\nu',\nu} - p_\kappa \delta_{\nu',\nu}] C_{\nu,\kappa} = 0 \qquad (27)$$

This set of secular equations differs greatly from the usual secular equations that are obtained when one diagonalizes the Hamiltonian of a quantum mechanical system. The kinetic energy term has disappeared, and the term representing nuclear attraction is diagonal. Furthermore, the eigenvalues are not energies but values of the parameter p_κ, which is related to the energy through equations (16) and (19). By solving the set of secular equations (27), one obtains a spectrum of p_κ values, and hence also a spectrum of energies $E_\kappa = -p_\kappa^2/2$. The lowest values of p_κ correspond to the most tightly bound states. From (18) we can see that this parameter can be interpreted as a “scaling parameter”, since the effective charges Q_ν that characterize the configurations are proportional to p_κ. Thus the basis set adjusts automatically to the energy of the state. For the lowest energy states, the basis functions are localized near the nucleus, while for highly excited states, they are diffuse. By solving the set of secular equations, one obtains simultaneously the energy spectrum and the optimal basis set for each state. This characteristic of the generalized Sturmian method makes it very well suited to the rapid calculation of large numbers of excited states of few-electron atoms, as is illustrated in Tables 1−5.

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