

Statistical Approximation for Exchange in Self-Consistent-Field Calculations of the Ground State of Neutral Argon*

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Comparisons are made of the orbital functions for the ground state of neutral argon obtained by four self-consistent-field methods. Three of these four are the familiar Hartree method, the Hartree-Fock method, and the Hartree-Fock method with Slater's statistical exchange potential. The fourth method is the same as Slater's but uses an exchange potential two-thirds as large as his. The smaller exchange potential was found by Kohn and Sham and by ourselves in a derivation which differs somewhat from Slater's. The orbitals computed from this set of self-consistent-field equations are closer to the Hartree-Fock orbitals than are the Hartree orbitals or the Hartree-Fock-Slater orbitals.

A CONSIDERABLE simplification of the Hartree-Fock self-consistent-field equations may be obtained by making a statistical (Thomas-Fermi) approximation for the exchange terms. This was first shown by Slater¹ who approximated the exchange term in the single electron Hartree-Fock equations,

$$-\sum_j \int \varphi_j^*(\mathbf{r}') \varphi_j(\mathbf{r}') \frac{1}{|\mathbf{r}-\mathbf{r}'|} d^3r' \varphi_j(\mathbf{r}),$$

by

$$-\frac{3}{2\pi} [3\pi^2 \rho(\mathbf{r})]^{1/3} \varphi_j(\mathbf{r}),$$

where $\rho(\mathbf{r})$ is the density of electrons. Recently, Kohn

and Sham² have shown that, if the approximation is made at an earlier stage in the derivation of the self-consistent-field equations—namely, in the expression for the total exchange energy—an exchange potential two-thirds that of Slater's is obtained. We have obtained the same result independently.

TABLE II. Slater F and G integrals^a for the ground state of neutral argon. The HFS or HFS' value closest to the corresponding HF value is starred. All values are in rydbergs.

| | HF | HFS | HFS' | H |
|--------------|--------|--------|---------|--------|
| $F^0(1s,1s)$ | 21.842 | 21.918 | 21.768* | 21.903 |
| $F^0(1s,2s)$ | 6.182 | 6.212* | 6.110 | 6.011 |
| $F^0(1s,2p)$ | 6.768 | 6.877 | 6.742* | 6.655 |
| $F^0(1s,3s)$ | 1.835 | 1.922 | 1.868* | 1.831 |
| $F^0(1s,3p)$ | 1.618 | 1.738 | 1.638* | 1.527 |
| $F^0(2s,2s)$ | 4.392 | 4.410* | 4.334 | 4.253 |
| $F^0(2s,2p)$ | 4.585 | 4.627* | 4.537 | 4.462 |
| $F^0(2s,3s)$ | 1.679 | 1.742 | 1.693* | 1.653 |
| $F^0(2s,3p)$ | 1.486 | 1.576 | 1.489* | 1.387 |
| $F^0(2p,2p)$ | 4.878 | 4.955 | 4.845* | 4.779 |
| $F^0(2p,3s)$ | 1.694 | 1.762 | 1.712* | 1.672 |
| $F^0(2p,3p)$ | 1.502 | 1.598 | 1.508* | 1.406 |
| $F^0(3s,3s)$ | 1.271 | 1.318 | 1.275* | 1.239 |
| $F^0(3s,3p)$ | 1.173 | 1.233 | 1.170* | 1.101 |
| $F^0(3p,3p)$ | 1.093 | 1.161 | 1.184* | 0.997 |
| $F^2(2p,2p)$ | 2.277 | 2.314 | 2.256* | 2.224 |
| $F^2(2p,3p)$ | 0.238 | 0.270 | 0.248* | 0.221 |
| $F^2(3p,3p)$ | 0.544 | 0.577 | 0.531* | 0.482 |
| $G^0(1s,2s)$ | 0.531 | 0.533* | 0.519 | 0.526 |
| $G^0(1s,3s)$ | 0.049 | 0.057 | 0.055* | 0.057 |
| $G^0(2s,3s)$ | 0.092 | 0.104 | 0.101* | 0.112 |
| $G^0(2p,3p)$ | 0.088 | 0.105 | 0.097* | 0.104 |
| $G^1(1s,2p)$ | 1.013 | 1.058 | 1.019* | 0.962 |
| $G^1(1s,3p)$ | 0.070 | 0.087 | 0.079* | 0.069 |
| $G^1(2s,2p)$ | 2.704 | 2.713* | 2.667 | 2.620 |
| $G^1(2s,3p)$ | 0.068 | 0.080 | 0.074* | 0.071 |
| $G^1(2p,3s)$ | 0.115 | 0.130 | 0.127* | 0.131 |
| $G^1(3s,3p)$ | 0.771 | 0.811 | 0.759* | 0.698 |
| $G^2(2p,3p)$ | 0.089 | 0.106 | 0.098* | 0.092 |

TABLE I. I integrals,^a "binding energies,"^b eigenvalues,^c and total energies,^d for the ground state of neutral argon. The HFS or HFS' value closest to the corresponding HF value is starred. All values are in rydbergs.

| | HF | HFS | HFS' | H |
|--------------------|-----------|-----------|------------|-----------|
| $1s$ | | | | |
| I | -323.741 | -323.798 | -323.688* | -323.799 |
| ϵ | -237.212 | -235.660 | -237.356* | |
| E | -237.212 | -232.449* | -227.426 | -237.118 |
| $2s$ | | | | |
| I | -77.785 | -77.906* | -77.426 | -76.882 |
| ϵ | -24.643 | -23.802 | -24.690* | |
| E | -24.643 | -22.785* | -21.457 | -22.201 |
| $2p$ | | | | |
| I | -76.324 | -76.769 | -76.162* | -75.763 |
| ϵ | -19.142 | -18.251 | -19.215* | |
| E | -19.142 | -18.127* | -16.754 | -17.849 |
| $3s$ | | | | |
| I | -27.028 | -27.804 | -27.226* | -26.761 |
| ϵ | -2.554 | -2.293 | -2.584* | |
| E | -2.554 | -2.053* | -1.665 | -1.912 |
| $3p$ | | | | |
| I | -23.568 | -24.725 | -23.617* | -22.289 |
| ϵ | -1.182 | -0.946 | -1.207* | |
| E | -1.182 | -1.007* | -0.667 | -0.834 |
| E_{total} | -1053.613 | -1053.336 | -1053.569* | -1053.464 |

^a $I_i = (i | -\nabla^2 - 2z/r | i)$.
^b $\epsilon_i = (i | -\nabla^2 - 2z/r | i) + \sum_j [(ij | 2/r_{12} | ij) - (ij | 2/r_{12} | ji)]$.
^c The E_i are the eigenvalues of the one-electron equations.
^d $E_{\text{total}} = \sum_i (i | -\nabla^2 - 2z/r | i) + \sum_{ij} [(ij | 1/r_{12} | ij) - (ij | 1/r_{12} | ji)]$. This expression for the expected value of the total energy is not quite correct when the Hartree orbitals are used as they are not exactly orthogonal.

* Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ J. C. Slater, Phys. Rev. **81**, 385 (1951).

^a $F^k(nl, n'l') = 2 \int_0^\infty \int_0^\infty R_n(r) R_{n'}(r') (r)^k (r')^{k+1} r^2 r'^2 dr dr'$

and

$G^k(nl, n'l') = 2 \int_0^\infty \int_0^\infty R_n(r) R_{n'}(r) R_n(r') R_{n'}(r') (r)^k (r')^{k+1} r^2 r'^2 dr dr'$,

where the R_n 's are normalized radial wave functions and $r_i = \min(r, r')$, $r_o = \max(r, r')$. It should be noted that the lack of orthogonality of the Hartree orbitals means that the Hartree F 's and G 's must be used differently from the others and hence are not strictly comparable.

² W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

TABLE III. Radial charge distribution of neutral argon in the ground state. Values are given at every tenth point of the radial mesh. The HFS or HFS' value closest to the HF value is starred. Hartree values are not given as they were computed using a different mesh.

| R^a | HF | $4\pi R^2\rho(R)^b$ HFS | HFS' |
|--------|---------|----------------------------|---------|
| 0.0084 | 2.540 | 2.567 | 2.526* |
| 0.0169 | 7.507 | 7.582 | 7.464* |
| 0.0253 | 12.493 | 12.609 | 12.422* |
| 0.0338 | 16.457 | 16.596 | 16.366* |
| 0.0507 | 20.500 | 20.651 | 20.408* |
| 0.0676 | 20.572 | 20.730 | 20.518* |
| 0.0845 | 18.785 | 18.980 | 18.786* |
| 0.1013 | 16.720 | 16.988 | 16.770* |
| 0.1351 | 14.521 | 14.986 | 14.609* |
| 0.1689 | 15.296 | 15.923 | 15.345* |
| 0.2027 | 17.508 | 18.194 | 17.482* |
| 0.2365 | 19.653 | 20.287 | 19.551* |
| 0.3040 | 21.104 | 21.426 | 20.922* |
| 0.3716 | 18.817 | 18.789* | 18.662 |
| 0.4392 | 14.809 | 14.579 | 14.744* |
| 0.5067 | 10.830 | 10.582 | 10.864* |
| 0.6419 | 5.753 | 5.799* | 5.903 |
| 0.7770 | 4.356 | 4.725 | 4.496* |
| 0.9121 | 4.702 | 5.223 | 4.772* |
| 1.0472 | 5.359 | 5.871 | 5.355* |
| 1.3175 | 5.701 | 5.956 | 5.603* |
| 1.5878 | 4.830 | 4.806* | 4.720 |
| 1.8580 | 3.600 | 3.409 | 3.523* |
| 2.1283 | 2.496 | 2.247 | 2.464* |
| 2.6688 | 1.073 | 0.8681 | 1.098* |
| 3.2093 | 0.4274 | 0.3101 | 0.4641* |
| 3.7498 | 0.1638 | 0.1067 | 0.1921* |
| 4.2903 | 0.0613 | 0.0361 | 0.0789* |
| 5.3714 | 0.00815 | 0.00403* | 0.01315 |
| 6.4524 | 0.00103 | 0.00045* | 0.00218 |

^a The unit of length is the Bohr radius.

^b $\int_0^\infty 4\pi R^2\rho(R) dR = Z = 18$.

It is our purpose here to compare the orbital functions obtained for atoms using the smaller statistical exchange potential (HFS') with those obtained from the conventional Hartree³ (H), Hartree-Fock⁴ (HF), and Hartree-Fock-Slater⁵ (HFS) equations. Rather than compare the radial functions directly we have chosen to compare various quantities which are commonly computed from the orbitals. We regard the Hartree-Fock orbitals as the standard by which the others are to be judged.

The results of our calculations for neutral argon in the ground state appear in Tables I-V. They clearly favor the use of the smaller exchange potential. Only the eigenvalues of the HFS' equations are consistently

³ D. R. Hartree, *The Calculation of Atomic Structures* (John Wiley & Sons, Inc., New York, 1957). The equations for the Hartree computer program and many of the numerical methods in all the programs are from this book.

⁴ J. C. Slater, *Quantum Theory of Atomic Structure* (McGraw-Hill Book Company, Inc., New York, 1960). The HF computer program is based primarily on this book.

⁵ F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963). Most of the HFS and HFS' calculations were done with computer programs based on the one developed by Herman and Skillman.

“poorer” than those of the HS or HF equations. This means the HFS' orbitals may be expected to be poor at large radii. Despite this expectation they give better estimates of $\langle r^4 \rangle$ and $\langle r^6 \rangle$ than the HFS or H orbitals in most cases.

One small difference between our calculations with the statistical exchange potential and many of those in literature should be pointed out. The results quoted here, except in Table V, are without the modification of the potential at large distances used by Latter,⁶ Herman and Skillman,⁵ and ourselves in an earlier paper.⁷ Calculations made with and without the “tail” correction indicate that it slightly improves the HFS results and makes the HFS' ones slightly worse. None of our conclusions are affected when calculations with it are used instead of the present ones.⁸

TABLE IV. Expectation values of powers of the radius for neutral argon orbitals. The unit of length is the Bohr radius. The HFS or HFS' value closest to the HF value is starred.

| | | HF | HFS | HFS' | H |
|----------|--------------------------|------------------------|-------------------------|--------------------------|------------------------|
| 1s | $\langle r^{-2} \rangle$ | 6.149×10^2 | 6.252×10^2 | $6.181 \times 10^{2*}$ | 6.229×10^2 |
| | $\langle r^{-1} \rangle$ | 1.755×10^1 | 1.761×10^1 | $1.750 \times 10^{1*}$ | 1.759×10^1 |
| | $\langle r \rangle$ | 8.610×10^{-2} | $8.581 \times 10^{-2*}$ | 8.642×10^{-2} | 8.579×10^{-2} |
| | $\langle r^2 \rangle$ | 9.959×10^{-3} | 9.888×10^{-3} | $10.033 \times 10^{-3*}$ | 9.866×10^{-3} |
| | $\langle r^4 \rangle$ | 2.559×10^{-4} | 2.509×10^{-4} | $2.586 \times 10^{-4*}$ | 2.481×10^{-4} |
| | $\langle r^6 \rangle$ | 1.274×10^{-5} | 1.214×10^{-5} | $1.272 \times 10^{-5*}$ | 1.185×10^{-5} |
| 2s | $\langle r^{-2} \rangle$ | 5.148×10^1 | 5.264×10^1 | $5.096 \times 10^{1*}$ | 4.937×10^1 |
| | $\langle r^{-1} \rangle$ | 3.555 | 3.575* | 3.511 | 3.446 |
| | $\langle r \rangle$ | 4.123×10^{-1} | $4.108 \times 10^{-1*}$ | 4.181×10^{-1} | 4.258×10^{-1} |
| | $\langle r^2 \rangle$ | 2.012×10^{-1} | $2.000 \times 10^{-1*}$ | 2.075×10^{-1} | 2.150×10^{-1} |
| | $\langle r^4 \rangle$ | 7.195×10^{-2} | $7.136 \times 10^{-2*}$ | 7.713×10^{-2} | 8.234×10^{-2} |
| | $\langle r^6 \rangle$ | 4.188×10^{-2} | $4.122 \times 10^{-2*}$ | 4.653×10^{-2} | 5.068×10^{-2} |
| 2p | $\langle r^{-3} \rangle$ | 1.244×10^2 | 1.309×10^2 | $1.244 \times 10^{2*}$ | 1.189×10^2 |
| | $\langle r^{-2} \rangle$ | 1.653×10^1 | 1.710×10^1 | $1.647 \times 10^{1*}$ | 1.600×10^1 |
| | $\langle r^{-1} \rangle$ | 3.450 | 3.508 | 3.438* | 3.389 |
| | $\langle r \rangle$ | 3.753×10^{-1} | 3.697×10^{-1} | $3.784 \times 10^{-1*}$ | 3.834×10^{-1} |
| | $\langle r^2 \rangle$ | 1.743×10^{-1} | 1.696×10^{-1} | $1.780 \times 10^{-1*}$ | 1.824×10^{-1} |
| | $\langle r^4 \rangle$ | 6.333×10^{-2} | $6.045 \times 10^{-2*}$ | 6.708×10^{-2} | 6.971×10^{-2} |
| 3s | $\langle r^{-3} \rangle$ | 4.058×10^{-2} | $3.814 \times 10^{-2*}$ | 4.485×10^{-2} | 4.662×10^{-2} |
| | $\langle r^{-2} \rangle$ | 5.365 | 6.266 | 5.977* | 5.903 |
| | $\langle r^{-1} \rangle$ | 9.621×10^{-1} | 10.135×10^{-1} | $9.843 \times 10^{-1*}$ | 9.656×10^{-1} |
| | $\langle r \rangle$ | 1.422 | 1.370 | 1.418* | 1.458 |
| | $\langle r^2 \rangle$ | 2.350 | 2.179 | 2.348* | 2.485 |
| | $\langle r^4 \rangle$ | 9.393 | 7.973 | 9.518* | 10.637 |
| 3p | $\langle r^{-3} \rangle$ | 6.023×10^1 | 4.611×10^1 | $6.287 \times 10^{1*}$ | 7.393×10^1 |
| | $\langle r^{-2} \rangle$ | 8.972 | 11.223 | 9.979* | 8.742 |
| | $\langle r^{-1} \rangle$ | 1.473 | 1.741 | 1.576* | 1.416 |
| | $\langle r \rangle$ | 8.140×10^{-1} | 8.752×10^{-1} | $8.246 \times 10^{-1*}$ | 7.683×10^{-1} |
| | $\langle r^2 \rangle$ | 1.663 | 1.564 | 1.682* | 1.832 |
| | $\langle r^4 \rangle$ | 3.311 | 2.918 | 3.435* | 4.110 |
| Σ | $\langle r^{-2} \rangle$ | 2.091×10^1 | 1.595×10^1 | $2.382 \times 10^{1*}$ | 3.488×10^1 |
| | $\langle r^{-1} \rangle$ | 2.302×10^2 | 1.506×10^2 | $3.054 \times 10^{2*}$ | 5.451×10^2 |
| | $\langle r \rangle$ | 1.451×10^3 | 1.481×10^3 | $1.458 \times 10^{3*}$ | 1.461×10^3 |
| | $\langle r^2 \rangle$ | 6.973×10^1 | 7.070×10^1 | $6.957 \times 10^{1*}$ | 6.895×10^1 |
| | $\langle r^4 \rangle$ | 1.607×10^1 | 1.533×10^1 | $1.621 \times 10^{1*}$ | 1.723×10^1 |
| | $\langle r^6 \rangle$ | 2.603×10^1 | 2.331×10^1 | $2.681 \times 10^{1*}$ | 3.117×10^1 |
| Σ | $\langle r^{-2} \rangle$ | 1.448×10^2 | 1.122×10^2 | $1.625 \times 10^{2*}$ | 2.312×10^2 |
| | $\langle r^{-1} \rangle$ | 1.502×10^3 | 0.996×10^3 | $1.958 \times 10^{3*}$ | 3.419×10^3 |
| | $\langle r \rangle$ | 1.451×10^3 | 1.481×10^3 | $1.458 \times 10^{3*}$ | 1.461×10^3 |
| | $\langle r^2 \rangle$ | 6.973×10^1 | 7.070×10^1 | $6.957 \times 10^{1*}$ | 6.895×10^1 |
| | $\langle r^4 \rangle$ | 1.607×10^1 | 1.533×10^1 | $1.621 \times 10^{1*}$ | 1.723×10^1 |
| | $\langle r^6 \rangle$ | 2.603×10^1 | 2.331×10^1 | $2.681 \times 10^{1*}$ | 3.117×10^1 |

⁶ R. Latter, Phys. Rev. **99**, 510 (1955).

⁷ D. Liberman, J. T. Waber, and D. T. Cromer, Phys. Rev. **137**, A27 (1965).

⁸ We are indebted to L. J. Sham for the suggestion that the tail correction, which was used in our earlier calculations, be removed.

TABLE V. X-ray scattering factors. The HFS or HFS' value closest to the HF value is starred. $s = \sin\theta/\lambda$ is in units of \AA^{-1} .

| s | HF ^a | HFS ^b | HFS' | H |
|------|-----------------|------------------|--------|-------|
| 0.00 | 18.00 | 18.00 | 18.00 | 18.00 |
| 0.05 | 17.54 | 17.57* | 17.49 | 17.45 |
| 0.10 | 16.30 | 16.41* | 16.16 | 16.03 |
| 0.15 | 14.65 | 14.84 | 14.47* | 14.24 |
| 0.20 | 12.93 | 13.20 | 12.80* | 12.51 |
| 0.25 | 11.42 | 11.70 | 11.35* | 11.06 |
| 0.30 | 10.20 | 10.45 | 10.18* | 9.93 |
| 0.35 | 9.25 | 9.46 | 9.27* | 9.07 |
| 0.40 | 8.54 | 8.70 | 8.57* | 8.43 |
| 0.50 | 7.56 | 7.64 | 7.57* | 7.52 |
| 0.60 | 6.86 | 6.91 | 6.84* | 6.83 |
| 0.70 | 6.23 | 6.30 | 6.22* | 6.19 |
| 0.80 | 5.61 | 5.70 | 5.60* | 5.55 |
| 0.90 | 5.01 | 5.11 | 5.00* | 4.93 |
| 1.00 | 4.43 | 4.54 | 4.42* | 4.34 |
| 1.10 | 3.90 | 4.01 | 3.89* | 3.82 |
| 1.20 | 3.43 | 3.54 | 3.43* | 3.35 |
| 1.30 | 3.03 | 3.13 | 3.03* | 2.96 |

^a *International Tables for X-ray Crystallography* (Kynoch Press, Birmingham, England, 1962), Vol. 3, p. 204.

^b H. P. Hanson *et al.*, *Acta Cryst.* **17**, 1040 (1964). These scattering factors and our HFS' scattering factors were computed from orbitals whose one-electron equations used a potential that was modified at large radii. Cf. F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963), pp. 1-8.

We also want to call attention to a related paper by Lindgren.⁹ He uses an exchange potential with three parameters which are adjusted to minimize the expected value of the total energy.

The statistical approximation may be used for calculating the effects of correlation as well as exchange.² Calculations including correlation have been made by Tong and Sham.¹⁰

Note added in proof. It has been pointed out by Slater¹¹ that Gaspar¹² suggested the use of the smaller exchange potential and has done a number of calculations with it. Unfortunately he used a charge density based on the one obtained by the Thomas-Fermi method. The inaccuracies introduced thereby are similar in magnitude to the differences between the various self-consistent calculations reported here.

⁹ I. Lindgren, *Arkiv. Fys.* (to be published). We are indebted to Dr. Lindgren for a copy of his report prior to publication.

¹⁰ B. Y. Tong and L. J. Sham, preceding paper *Phys. Rev.* **144**, 1 (1966).

¹¹ J. C. Slater, Massachusetts Institute of Technology, Solid-State and Molecular Theory Group, Quarterly Progress Report, No. 58, 1965 (unpublished).

¹² R. Gaspar, *Acta Phys. Hung.* **3**, 263 (1954), and subsequent publications.

Lifetime, Coherence Narrowing, and Hyperfine Structure of the $(6s^26p7s)^3P_1^0$ State in Lead*

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The lifetime and hyperfine structure of the $(6s^26p7s)^3P_1^0$ excited state in lead have been investigated by the technique of level-crossing spectroscopy. The lifetime of the state was determined to be $\tau = 5.75(20) \times 10^{-9}$ sec. The linewidth of the level-crossing signal was observed to saturate at a "coherence-narrowed" value in agreement with theoretical predictions. The magnitude of this narrowing corresponds to a 27(3)% branching ratio to the ground state. When the branching ratio and the lifetime are combined with the $f(3639 \text{ \AA}) : f(4058 \text{ \AA})$ oscillator-strength ratio measured by Khokhlov, one may infer the absolute f values: $f(2833 \text{ \AA}) = 0.169(17)$, $f(3639 \text{ \AA}) = 0.040(8)$, $f(4058 \text{ \AA}) = 0.155(31)$. The high-field level crossing in the hyperfine structure of the first $^3P_1^0$ state of Pb^{207} was found to occur at a magnetic field $H_0 = 4661.1(4)$ G, which corresponds to a magnetic-dipole-interaction constant $A = 8.811(17) \times 10^8$ cps.

I. INTRODUCTION

IN this paper we report on an experimental determination of the lifetime, coherence narrowing, and hyperfine structure of the $(6s^26p7s)^3P_1^0$ excited state in lead by level-crossing spectroscopy.

Precise values of the lifetime of an excited state of an atom and the related oscillator strengths (f values) are required by astrophysicists for studies of stellar struc-

ture and by plasma physicists for the determination of the properties of high-temperature plasmas. In addition, the experimental values provide a valuable check on approximation methods used to estimate atomic radiative properties. Knowledge of the oscillator strengths is needed in the calculation of emission and absorption probabilities, of resonant collision broadening of the optical line, of the collision broadening of level-crossing and double-resonance signals, and of the phenomenon of coherence narrowing. In the case of complex atoms, the observed lifetimes may be useful in determining the mixing of atomic states of different multiplicities.

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