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ATOMIC BINDING ENERGIES FROM A MODIFIED THOMAS-FERMI-DIRAC THEORY*

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ABSTRACT

A "quantum correction" of the statistical model of the atom has been obtained by modifying March and Plaskett's region of integration in the (n_r, ℓ), or quantum number, plane. Integrations over the plane lead, in the unmodified case, to the Thomas-Fermi density expression and energy equation. Integrations over the modified region have here been shown to produce a modified Thomas-Fermi expression for the electron density, and a correction to the kinetic energy. The latter correction shows a similarity to the Weizsäcker correction, but is smaller by a slowly changing factor of the order of ten. A modified Thomas-Fermi-Dirac equation has been derived by the standard variational procedure. Numerical solutions of the equation have been obtained which yield atomic binding energies in much better agreement with experimental values than those of the unmodified theory.

I. INTRODUCTION

The statistical model of the atom, first propounded by Fermi[1] and Thomas,[2] and extended notably by Dirac,[3] has proved a most useful approximation to the self-consistent field method in calculating electron distributions and fields in atoms. Because of its relative simplicity, it has found wide application as a means of predicting properties of free atoms and of solids.[4]

There has been considerable interest recently in extension of the statistical theory through the incorporation of "quantum corrections."[5] However, the inclusion of quantum effects, with the exception of the exchange energy, leads to severe complication of the theory and of the equations which must be solved by numerical means. There should be merit in a quantum correction which, though perhaps lacking a firm underlying basis, remains tractable while exhibiting the possibility of useful application.

Prompting at least some of the numerous modifications of the theory, including the present one, is the knowledge that the discrepancy is quite large between the observed total binding energy and that calculated, either with or without consideration of exchange effects. For illustration the total energies for the low Z elements, for which experimental values

Table I

have been obtained, are given in Table I. The Thomas-Fermi (TF) energies have been calculated according to the formula due essentially to Milne:[6]

$$W_{TF} = -20.92\ Z^{7/3} \text{ev}.$$

The Thomas-Fermi-Dirac (TFD) energies have been obtained from the paper of Cowan and Ashkin,[7] and the experimental values are from Moore.[8]

We shall show that a quantum correction can be derived by modification of a region of integration in the (n_r, ℓ), or quantum number, plane employed by March and Plaskett[9] in their derivation of the TF energy equation. A quantum-correction energy density is identified, and it is then possible to obtain a quantum-corrected TFD equation by the usual variational procedure. Numerical solutions of the equation yield atomic binding energies in very good agreement with experimental and Hartree values.

The discussion presumes zero temperature; a possible extension to nonzero temperature is outlined at the end of the paper.

II. A MODIFICATION OF MARCH AND PLASKETT'S INTEGRATIONS

For details of March and Plaskett's derivation the reader is referred to their paper. Briefly, they have shown that the sum of one-electron eigenvalues in a spherically symmetric potential is approximated in the TF method by an integral of the WKB eigenvalues over a particular region of the (n_r, ℓ) plane. We have used n_r to denote the radial quantum number, and ℓ is the orbital quantum number. The WKB eigenvalues are obtained as solutions of the equation[10]

$$2\int_{r_1^{\ell}(E)}^{r_2^{\ell}(E)} \left\{2m[E-V(r)-(\hbar^2/2m)(\ell+\overset{\text{Cancel}}{1/2})^2/r^2\right\}^{1/2} dr = (n_r+\overset{\text{Cancel}}{1/2})h, \qquad (1)$$

where $r_1^{\ell}(E)$ and $r_2^{\ell}(E)$ are the roots of

$$V(r) + (\hbar^2/2m)(\ell+\overset{\text{Cancel}}{1/2})^2/r^2 = E,$$

V(r) being potential energy. March and Plaskett have demonstrated that the statistical approximation to the sum of eigenvalues is given by the integral[11]

$$I = 2\int\int (2\ell+1)E(n_r,\ell)dn_r d\ell, \qquad (2)$$

where the number of states over which the sum is carried is written as

$$N = 2\int\int (2\ell+1)dn_r d\ell. \qquad (3)$$

The region of integration in Eqs. (2) and (3) is bounded by $n_r = -1/2$, $\ell = -1/2$, and $E(n_r,\ell) = E'$. The Fermi energy, E', is chosen so that Eq. (3) gives the total number of states being considered, the N electrons occupying the N lowest states at zero temperature. $E(n_r,\ell)$ is the expression for the WKB eigenvalues considered as functions of continuous variables. By manipulating Eqs. (1), (2), and (3), March and Plaskett have derived the TF energy equation:

$$I = \int \left(\frac{3}{5}\frac{P^2}{2m} + V\right)\frac{8\pi P^3}{3h^3}\, 4\pi r^2 dr \tag{4}$$

and the expression

$$N = \int \frac{8\pi P^3}{3h^3}\, 4\pi r^2 dr, \tag{5}$$

the integrals being taken between the roots of $E' = V(r)$. From Eq. (5), the TF density is identified as

$$\rho = 8\pi P^3/3h^3,$$

where $P = [2m(E'-V)]^{1/2}$ is the Fermi momentum.

When the potential is known, the evaluation of the TF approximation to the sum of eigenvalues is simply effected by the use of Eqs. (2) and (3). In the particular case of a Coulomb field, Scott's correction[12] to the atomic binding energy is obtained in comparing the approximate sum with the correct sum. Thus, in atomic units the WKB expression for the eigenvalues in a Coulomb field is

$$E = -Z^2/2(n_r+\ell+1)^2,$$

identical with that obtained by solving Schrödinger's equation. We introduce for convenience

$$\begin{aligned}\alpha &= (-Z^2/2E')^{1/2} \qquad (6)\\ &= (n_r+\ell+1)_{\text{outer boundary}},\end{aligned}$$

the subscript referring to the outer boundary of the region of integration in the (n_r,ℓ) plane.

We now have, from Eq. (3),

$$\begin{aligned}N &= 2\int_{\ell=-1/2}^{n_r+\ell+1=\alpha}\int_{n_r=-1/2}(2\ell+1)dn_r d\ell\\ &= 2\alpha^3/3. \qquad (7)\end{aligned}$$

If levels are filled from n = 1 to n = ν, where n is defined by $n = n_r+\ell+1$, then the number of states must equal

$$\sum_1^{\nu} 2n^2 = \nu(\nu+1)(2\nu+1)/3. \qquad (8)$$

Therefore, equating Eqs. (7) and (8),

$$\alpha = [\nu(\nu+1)(2\nu+1)/2]^{1/3}.$$

Carrying out the integration of Eq. (2) in a similar manner and substituting for α, we get

$$I = -Z^2[\nu(\nu+1)(2\nu+1)/2]^{1/3}. \qquad (9)$$

Scott's correction, $Z^2/2$, is then obtained by subtracting Eq. (9) from the correct sum of eigenvalues, $-Z^2\nu$, and letting ν tend to infinity.

In application to the statistical atom, the sum of one-electron eigenvalues is not the total energy of the electron distribution, since the electron-electron potential energy is included twice in the summation. However, the over-estimation of the atomic binding energies is caused by the large magnitude of the electron-nuclear potential energy resulting from the infinite density of electrons which the theory predicts at the nucleus. Since correction of the electron-electron potential energy is thus of minor importance, we might expect to achieve a significant improvement in binding energy by correcting, in some manner, the sum of eigenvalues.

To pursue this end, let us consider the following modification of the available electron states. Let us change the lower limit of ℓ and the value of α so that the correct sum of eigenvalues results, again in the case of the Coulomb field, when integrations such as the preceding are performed over the modified region. We shall denote the lower limit of ℓ by ℓ_{min}, which is, in general, now different from -1/2. It is convenient also to introduce the quantity $a = \ell_{min}+1/2$, which we shall call the "modification factor." An evaluation of a and α for the K shell follows.

To include two states in the region of integration, we require that

$$2 = 2\int_{\ell=a_K-1/2}^{n_r+\ell+1=\alpha_K}\int_{n_r=-1/2}(2\ell+1)\,dn_r\,d\ell$$

$$= 2(\alpha_K{}^3-3\alpha_K a_K{}^2+2a_K{}^3)/3.$$

The condition that the total energy of the two K-electrons be the correct value yields

$$2(-Z^2/2) = 2\int_{\ell=a_K-1/2}^{n_r+\ell+1=\alpha_K}\int_{n_r=-1/2}(2\ell+1)[-Z^2/2(n_r+\ell+1)^2]\,dn_r\,d\ell$$

$$= -Z^2(\alpha_K{}^2-2\alpha_K a_K+a_K{}^2)/\alpha_K.$$

The pertinent solution of these two equations is $a_K = 0.26679643$, $\alpha_K = 1.4856820$.

Putting $\nu = 2$, a similar calculation for the ten states of lowest energy results in the values $a_L = 0.25928018$, $\alpha_L = 2.4915790$. Further, we can consider Eq. (8) to represent the total number of states for non-integral values of ν; corresponding values of a and α can then be found.

It is not difficult to show that as the number of filled shells becomes very large, ℓ_{min} tends toward the unmodified TF value of -1/2, and that as the region of integration goes to zero, $\alpha = a = 6^{-1/2} = 0.40824829$.

If one performs the integrations indicated in Eqs. (2) and (3) with the general value ℓ_{min} replacing -1/2 as the lower limit of ℓ, the result is a modified TF energy equation and a modified formula for the density. Specifically, we obtain in place of Eqs. (4) and (5),

$$I = \int_{r_1}^{r_2} \left[\frac{3}{5}\frac{p^2}{2m} + V + \frac{\hbar^2}{5m}\frac{a^2}{r^2}\right]\rho \, 4\pi r^2 dr, \tag{10}$$

and

$$N = \int_{r_1}^{r_2} \frac{8\pi}{3h^3}\left[2m(E'-V-\frac{\hbar^2}{2m}\frac{a^2}{r^2})\right]^{3/2} 4\pi r^2 dr. \tag{11}$$

The electron density in Eq. (10) has been identified from Eq. (11). That is,

$$\rho = (8\pi/3h^3)\left[2m(E'-V-\frac{\hbar^2}{2m}\frac{a^2}{r^2})\right]^{3/2} \tag{12}$$

in the region specified by the limits on Eqs. (10) and (11). These limits are the roots[13] of

$$V + (\hbar^2/2m)a^2/r^2 = E'. \tag{13}$$

Obviously what we have done is to eliminate states with orbital angular momentum between zero and a cut-off value $L_c = a\hbar$. Corresponding to L_c at every radial distance is a minimum value of allowed linear momentum, or more specifically, the lowest allowed magnitude of a momentum vector having no radial component. Calling this linear cut-off momentum $p_c = a\hbar/r$ allows us to write the density as

$$\rho = (8\pi/3h^3)(P^2-p_c^2)^{3/2} \tag{14}$$

At radial distances less than r_1, momenta are prohibited over the entire range from zero to P, so the electron density vanishes.

By integrating over the region in momentum space which remains after elimination of a circularly cylindrical portion oriented along the radial momentum axis, we obtain

$$(p^2)_{av} = 3P^2/5 + 2p_c^2/5.$$

The kinetic energy density is therefore given by

$$U_k = (1/2m)(3P^2/5+2p_c^2/5)\rho. \tag{15}$$

Using Eq. (14) we can write Eq. (15) as

$$U_k = c_f\rho^{5/3} + (c_q/r^2)\rho, \tag{16}$$

where $c_f = (3\hbar^2/10m)(3\pi^2)^{2/3}$ and $c_q = (\hbar^2/2m)a^2$. The first term on the right side of Eq. (16) is the usual expression for the Fermi kinetic energy density. The second term is a correction which we shall call the quantum-correction energy density, U_q.

III. MODIFIED TF AND TFD EQUATIONS

A modified TF equation follows immediately from Eq. (12) and Poisson's equation. Following the usual procedure, the equation for the TF potential function ϕ, defined by $Ze^2\phi = (E'-V)r$, is obtained as

$$\begin{aligned} \phi'' &= (4x/3\pi Z)(2Z\phi/x - a^2/x^2)^{3/2}, && x \geqq x_1, \\ &= 0, && x < x_1. \end{aligned} \tag{17}$$

Here x is distance measured in units of the first Bohr radius for hydrogen. The boundary conditions are the same as for the unmodified equation:

$$\phi(0) = 1$$

$$x_2\phi'(x_2) = \phi(x_2).$$

A modified TFD equation can be derived by the variation technique employed originally by Jensen[14] and recently by Tomishima[15] in his inclusion of correlation effects. The total energy density of the distribution is written as

$$U = c_f\rho^{5/3} - c_{ex}\rho^{4/3} - e(V^n+V^e/2)\rho + (c_q/r^2)\rho,$$

where $c_{ex} = (3/4)(3/\pi)^{1/3} e^2$, and v^n and v^e are the potentials due to the nucleus and the electrons, respectively. Minimization of the total energy integral leads to the equation

$$(5c_f/3)\rho^{2/3} - (4c_{ex}/3)\rho^{1/3} + c_q/r^2 + V - E' = 0,$$

which yields

$$\rho = \sigma_o[\tau_o+(E'-V-c_q/r^2+\tau_o^2)^{1/2}]^3, \qquad r \geqq r_1.$$

Here $\sigma_o = (3/5c_f)^{3/2}$ and $\tau_o = (4c_{ex}{}^2/15c_f)^{1/2}$. The positive sign of the square root is chosen so that the density agrees with the TF expression if the exchange, represented by τ_o, is neglected. We again choose r_1 as the radius at which $(E'-V-c_q/r^2)$ vanishes. At $r = r_1$ the density is therefore $8\sigma_o\tau_o^3$, while again we set $\rho = 0$ for $r < r_1$. Understanding ϕ now to represent the TFD potential function, defined by $Ze^2\phi = (E'-V+\tau_o^2)r$, we are led to the modified TFD equation:

$$\begin{aligned}\phi'' &= (4x/3\pi^4Z)[1+\pi(2Z\phi/x-a^2/x^2)^{1/2}]^3, && x \geqq x_1,\\ &= 0, && x < x_1.\end{aligned} \qquad (18)$$

The same boundary conditions apply as in the previous case.

In performing integrations of Eq. (17) or (18), the modification factor, a, must be specified. As we have previously implied, we have made the approximation that the electrons are moving in a pure Coulomb field in order to simplify the numerical work. Under this approximation ϕ is a linear function of distance from the nucleus, and Eq. (6) becomes $\alpha = (-Z/2\phi'_0)^{1/2}$, where ϕ'_0 is the initial slope of the potential function with respect to x. With α a known quantity, a can then be found.

It should be pointed out that a and α thus computed will not correspond to the values which we would obtain for an element of atomic number Z by simply considering Z electrons to be moving in a pure Coulomb field. In fact, it develops that the modification factor for the isolated hydrogen atom is to be computed on the basis of about 0.10 electrons in such a field, and for the isolated atom with Z = 100 we still get only about 11 particles.

The Coulomb approximation is certainly suggested by the success of Scott's correction, to which we have previously referred. The consistency of the approximation with the results obtained from integrating Eq. (18) has been examined in some detail and found to be quite good. The alternative to making this simplification is by an iterative process to obtain a modified electron distribution in which the calculated sum of one-electron eigenvalues is "self-consistent." That is, we can calculate numerically the eigenvalues in the shielded potential of the nucleus, say by the WKB method; these are then summed. We can also evaluate the sum of eigenvalues by adding an amount of energy equal to the electron-electron potential energy to the total energy of the electron distribution as calculated by the statistical method. We can then demand agreement of the two sums.

IV. RESULTS

Numerical integrations of Eq. (18) have been performed[16] for several atomic species using the Los Alamos IBM type 704 digital computers. Some of the results are summarized here.

Table II

In Table II the calculated total energies of the isolated atoms may be compared with the energies computed according to a formula due to Foldy,[17] which is based on results of Hartree calculations. With the exception of Z = 1, the agreement is nowhere worse than to within 1.5 percent, with most discrepancies well under one percent.

Several comparisons are of interest. Among these are the values computed for x_1, as compared with those obtained by Golden.[5] In Golden's paper the information is given from which this quantity, the inner radius at which the electron density vanishes, may be calculated for the ground states of the atoms with Z = 1, 2, and 8. The comparison is made in Fig. 1. From the curves it is apparent that the values obtained in the present calculations range from about three times those of Golden at low Z to about 1.5 times his (projected) values at high Z.

Fig. 1

We have also compared the radial behavior of the quantum-correction energy density with the Weizsäcker inhomogeneity energy density.[18] Using the calculated electron distribution for the isolated copper atom for evaluation of these quantities, we have plotted in Fig. 2 the Weizsäcker energy density U_i (multiplied by a rather arbitrary factor of 1/10), the quantum-correction energy density U_q, and the corresponding radially weighted quantity $U_q x^2$. It is seen that the former two curves differ by

Fig. 2

less than an order of magnitude over the spatial region in which $U_q x^2$, which measures the contribution to the quantum-correction energy at a given radius, varies by several orders of magnitude. This is of interest because the Weizsäcker energy term is considered by several authors[19] to be too large by a factor of 9. Very near the nucleus the correspondence breaks down, however, since the Weizsäcker energy possesses a zero at the radius at which the electron density has its maximum value.

V. EXTENSION TO NONZERO TEMPERATURE

Extension to temperature other than zero is particularly simple if one neglects exchange effects. From Eq. (14) the number of states per unit volume with momenta between p and p + dp is evaluated as $(8\pi/h^3)(p^2-p_c^2)^{1/2}pdp$. Since the probability of occupation of the jth state, with energy $E_j = p_j^2/2m + V_j$, is given by $n_j = [\exp\beta(E_j-\mu)+1]^{-1}$, where $\beta = 1/kT$ and μ is the chemical potential, we have as the formula for the electron density

$$\rho = (8\pi/h^3)\int_{p_c}^{\infty} [\exp\beta(E_j-\mu)+1]^{-1}(p^2-p_c^2)^{1/2}pdp. \qquad (19)$$

Introducing a new momentum variable defined by $(p')^2 = p^2 - p_c^2$, Eq. (19) can be written in terms of the "Fermi-Dirac function"[20] $F_{1/2}(\zeta)$. We then have

$$\rho = (4\pi/h^3)(2m/\beta)^{3/2} F_{1/2}(\zeta),$$

the variable of integration in the Fermi-Dirac function being defined by $y = \beta(p')^2/2m$, and ζ standing for[21] $-\beta(p_c{}^2/2m + V - \mu)$. The equation for the potential function could now be set up and solved exactly as described, for example, by Latter.[22] The solutions would differ from those obtained by Latter chiefly in the vicinity of $r = 0$. In the unmodified case we have $\zeta = -\beta(V-\mu)$, and as the nucleus is approached $\zeta \rightarrow \infty$ and $F_{1/2}(\zeta) \rightarrow \infty$ as $\zeta^{3/2}$. Under the modification, with ζ including the term in p_c, $\zeta \rightarrow -\infty$ and $F_{1/2}(\zeta) \rightarrow 0$ as e^{ζ}.

The evaluation of p_c, or equivalently of the modification factor, should proceed just as in the zero temperature case.

TABLE I. Comparison of the total atomic binding energies on the Thomas-Fermi and Thomas-Fermi-Dirac models with experimental values.

Z	$-W_{TF}$(ev)	$-W_{TFD}$(ev)	$-W_{exp}$(ev)
1	20.92	28.07	13.60
2	105.4	126.7	78.98
3	271.5	312.4	203.4
4	531.3	596.3	399.0
5	894.3	987.5	670.8
6	1369.	1494.	1030.
7	1961.	2122.	1486.
8	2678.	2878.	2043.

TABLE II. Total atomic binding energies from the modified theory compared with Foldy's values.

Z	$-W_{\text{mod TFD}}$(ev)	$-W_{\text{Foldy}}$(ev)
1	15.50	13.60
2	77.52	78.69
3	201.8	202.2
4	399.4	400.6
5	679.1	677.3
6	1049.	1042.
7	1515.	1507.
8	2084.	2070.
10	3548.	3538.
20	$1.856 \cdot 10^4$	$1.834 \cdot 10^4$
30	4.881	4.815
40	9.682	9.597
50	$1.646 \cdot 10^5$	$1.638 \cdot 10^5$
60	2.539	2.534
70	3.662	3.666
80	5.030	5.053
90	6.651	6.710
100	8.540	8.585

FIGURE LEGENDS

Figure 1.	The inner radius, below which $\rho = 0$, in atomic units.
Figure 2.	A comparison of the quantum-correction energy density and the Weizsäcker inhomogeneity energy density for the isolated copper atom.

FOOTNOTES

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FOOTNOTES (Cont.)

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[13] In application to compressed atoms, there is only one root of Eq. (13) between zero and the outer boundary of the neutral atom. This root is identified as r_1, and r_2 is then determined by the usual TF boundary condition.

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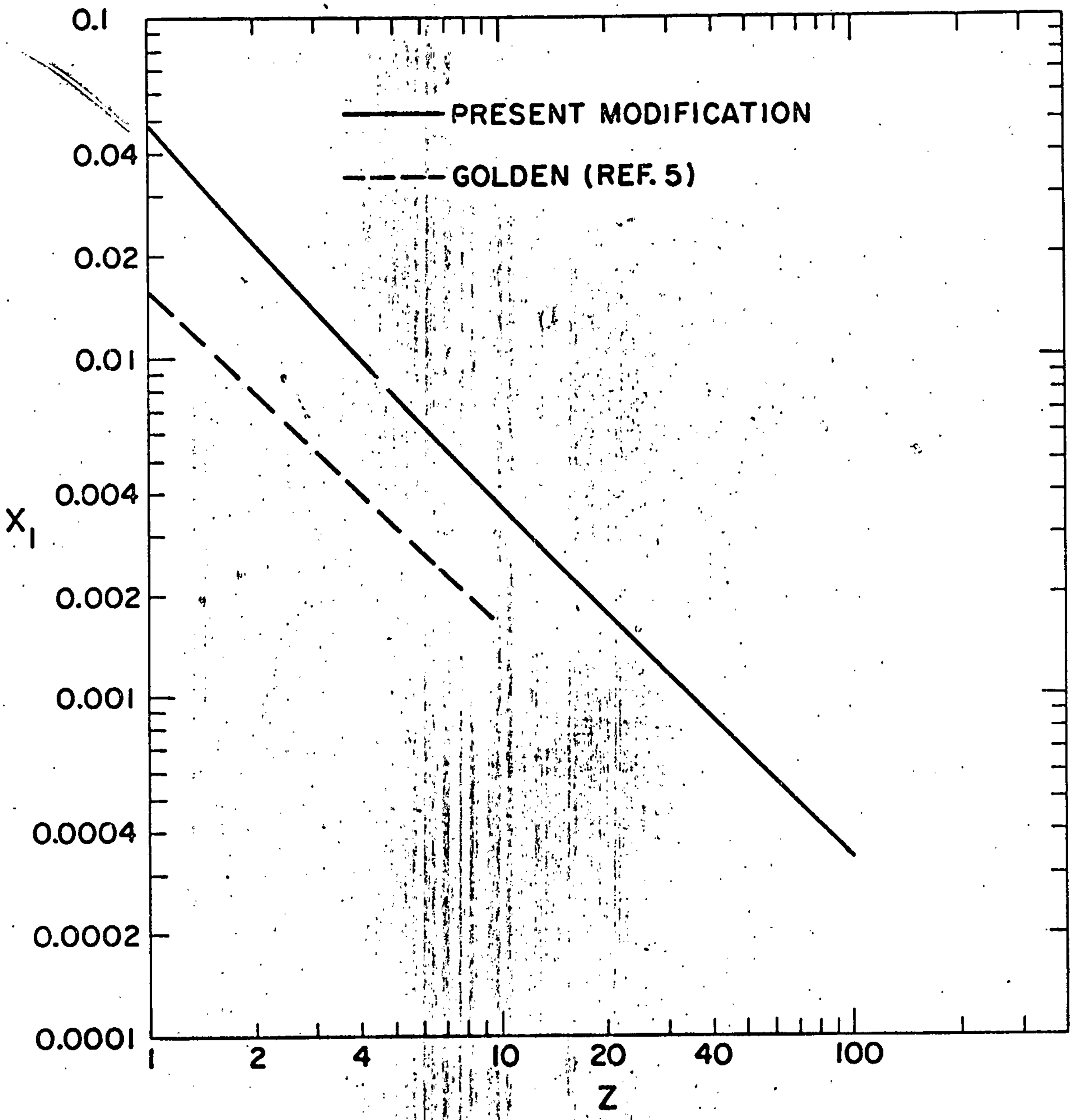
J. F. Barnes & R. D. Cowan

Fig. 1



J. F. Barnes & R. D. Cowan

Fig 2