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Inhomogeneous relativistic electron gas: Correlation potential

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The correlation energy of a relativistic electron gas based on a sum of ring diagrams including the transverse-photon contributions is numerically calculated. From this, a local, relativistic correlation potential is deduced which appears in the density-functional theory of inhomogeneous systems where relativistic effects make important contributions. Comparison with its nonrelativistic counterpart shows that the use of nonrelativistic results is very poor. The correlation contribution to the potential is found to be about 5% of the exchange for $\beta=2.5$ to about 2.5% for $\beta=1$ ($\beta=k_F/m$).

I. INTRODUCTION

In a recent paper, one of us (Rajagopal,[1] referred to as I henceforth) presented a relativistic generalization of the Hohenberg-Kohn-Sham theory of the inhomogeneous nonrelativistic electron gas. In a local scheme, using the exchange energy of a uniform relativistic electron gas, the exchange part of the effective potential was deduced (see also MacDonald and Vosko[2]). In this paper, we present the correlation part of the potential, based on a calculation of the ground-state energy of the uniform relativistic electron gas, with a ring-diagram sum approximation which is a generalization of the nonrelativistic result of Gell-Mann and Brueckner[3] (GB). There are several independent derivations of the properties of relativistic electron gas in the literature, all of which are equivalent; the reason for the existence of so many of these works is because this model problem is of great interest in many fields—nuclear physics (Chin[4]), astrophysics (Jancovici[5] and Akhieser and Peleteminskii[6]), many-electron physics (Fradkin[7] and Bowers *et al.*[8]), and plasma physics (Tsytovich[9] and Bezzerides and DuBois[10]).

The importance of relativistic effects, especially when heavy-atom systems are involved, in atomic, molecular, and solid-state physics has been known for a long time and only recently, because of sophisticated computing facilities and methods, actual calculations are being reported. For a review of these, one may refer to Pyykkö[11] and Desclaux.[12] In these applications, it is felt, as in the nonrelativistic theory, a self-consistent density-functional theory may be a useful scheme. An application of our relativistic theory with exchange only approximation has now been shown to yield significant improvement over the Dirac-Slater (Dirac equation with Slater $\rho^{1/3}$ potential) atomic structure calculations.[13] In the nonrelativistic formalism, it is known that correlation contribution to the potential is important and there are several schemes for setting up this contribution from the uniform nonrelativistic many-electron theory. For a review, one may see the article of Rajagopal.[14] In this paper, we present a correlation potential, based on an approximation scheme for calculating the ground-state energy of the electron-gas system. Unlike the nonrelativistic theory, we have two kinds of contributions arising from the longitudinal and transverse photon-electron interactions. The latter term is negligible in the nonrelativistic limit but makes significant contributions as relativistic effects become important. This was already evident in the exchange-energy and potential calculation presented in I. In the case of atomic systems with $Z > 50$, relativistic effects become significant. In the next section, a brief outline of the theory is presented, spelling out the approximations and the resulting renormalizations, etc., as well as the separate longitudinal and transverse contributions to the correlation energy and potential. The numerical results are displayed as figures and representative numerical values are given as tables in the third section. Comparison with the exchange contributions are also made. The calculations are made for values of electron densities appropriate to condensed-matter systems composed of atoms. In the final section, a summary of the results with concluding remarks are given.

II. THEORY

We use notations as in the book of Björken and Drell.[15] By a straightforward manipulation, we express the exchange-correlation energy per unit volume (Ω is the volume of the system) of a relativistic electron gas in terms of the matter current-correlation functions and the electromagnetic-field correlation functions

$$E_{\rm xc}^{\rm tot}=\frac{1}{\Omega}\int_0^e\frac{de_0}{e_0}\int d^3r_1 e_0\langle\bar{\psi}(1)\gamma_\mu\psi(1)A^\mu(1)\rangle_{e_0}$$

$$=\frac{i}{2\Omega}\int_0^{e^2}\frac{de_0^2}{e_0^2}\int d^3r_1 d^4 2 Q_{\nu\mu}(12)D^{\mu\nu}(21^+)$$

$$+\frac{1}{2\Omega}\int_0^{e^2}\frac{de_0^2}{e_0^2}\int d^3r_1 e_0\langle\bar{\psi}(1)\gamma_\nu\psi(1)\rangle\langle A^\nu(1)\rangle\ . \tag{1}$$

Here $Q_{\nu\mu}$ (12) is the exact matter current-current-correlation function and $D^{\mu\nu}$ is the electromagnetic-field correlation function which obeys the Maxwell field equations in the presence of matter. $\nu,\mu=0,1,2,3$ and the repeated indices imply sum over them. Using the Coulomb gauge and working to leading order in interaction strength, we obtain (S_F is the free-electron Feynman propagator) in momentum space:

$$Q_{\mu\nu}(k)\cong -4\pi e_0^2 i\int\frac{d^4p}{(2\pi)^4}\mathrm{tr}[\gamma_\nu S_F(k+p)\gamma_\mu S_F(p)]\ , \tag{2}$$

and

$$D^{00}(k)\cong\frac{1}{|\vec{k}|^2}\left[1-\frac{1}{|\vec{k}|^2}Q_{00}(k)\right]^{-1}\ , \tag{3a}$$

$$D^{0j}(k)=0\ , \tag{3b}$$

$$D^{ij}(k)=-\frac{(\delta^{ij}-k^ik^j/|\vec{k}|^2)}{(k_0^2-|\vec{k}|^2)}\left[1-\frac{1}{k_0^2-|\vec{k}|^2}Q_T(k)\right]^{-1}\ , \tag{3c}$$

with $(i,j=1,2,3)$.

Here $Q_T(k)$ is the transverse part of Q_{ij} obtained from (2) by the formula

$$Q_T(k)=\tfrac{1}{2}\left[\delta^{ij}-\frac{k^ik^j}{|\vec{k}|^2}\right]Q_{ij}(k)\ . \tag{3d}$$

Explicitly working out the indicated traces and performing the relevant integrals, we obtain finally the exchange-correlation energy, in a more familiar form:

$$E_{\rm xc}^{\rm rings}=\frac{1}{2\Omega}\int\frac{d^3q}{(2\pi)^3}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\ln\left[1+\frac{4\pi e^2}{q^2}Q_{00}(q,\omega)\right]+\frac{1}{2\Omega}\int_0^{e^2}\frac{de_0^2}{e_0^2}\int d^3r_1 e_0\langle\bar{\psi}\gamma_\nu\psi\rangle\langle A^\nu\rangle$$

$$+\frac{1}{\Omega}\int\frac{d^3q}{(2\pi)^3}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\ln\left[1-\frac{8\pi e^2}{q^2+\omega^2}Q_T(q;\omega)\right]\ . \tag{4}$$

Here we have already converted the frequency integral $\omega\to i\omega$. Using

$$S_F(k)=\left[\frac{\not{k}+m}{2E_k}\right]_{k_0=E_k}\left[\frac{1-n_F(k)}{k_0-E_k+i\eta}+\frac{n_F(k)}{k_0-E_k-i\eta}\right]-\left[\frac{\not{k}+m}{2E_k}\right]_{k_0=-E_k}\frac{1}{k_0+E_k-i\eta}\ , \tag{5}$$

where $E_k=(m^2+\vec{k}^2)^{1/2}$, we can then compute Q_{00},Q_T in a straightforward way. In Eq. (5) we have explicitly exhibited the fact that the positive energy states are occupied. In obtaining the expressions for the vari-

ous $Q_{\nu\mu}$ upon using Eq. (5) we obtain some terms involving no density (k_F), the Fermi momentum is related to density n via $k_F^3/3\pi^2=n$), which are just due to the pure quantum electrodynamical renormalizations, which we incorporate in our scheme by using physical charge and mass of the electron. We concentrate our attention on the "many-particle" aspect by examining only the density-dependent terms. With this observation, then, the relevant expressions for Q_{00} and Q_T that appear in Eq. (4) are

$$Q_{00}(q,\omega)\cong\int_{k<k_F}\frac{d^3k}{(2\pi)^3}\left[\frac{(E_{k+q}-E_k)[(E_{k+q}+E_k)^2-q^2]}{E_kE_{k+q}[(E_{k+q}-E_k)^2+\omega^2]}+\frac{(E_{k+q}+E_k)[E_{k+q}-E_k)^2-q^2]}{E_kE_{k+q}[(E_{k+q}+E_k)^2+\omega^2]}\right], \tag{6}$$

$$Q_T(q;\omega)\cong\int_{k<k_F}\frac{d^3k}{(2\pi)^3}\left[\frac{(E_{k+q}-E_k)[E_{k+q}E_k-(\vec{k}\cdot\vec{q})^2/q^2-(\vec{k}\cdot\vec{q})-m^2]}{E_kE_{k+q}[(E_{k+q}-E_k)^2+\omega^2]}\right.$$

$$\left.-\frac{(E_{k+q}+E_k)[E_{k+q}E_k+(\vec{k}\cdot\vec{q})^2/q^2+(\vec{k}\cdot\vec{q})+m^2]}{E_kE_{k+q}[(E_{k+q}+E_k)^2+\omega^2]}\right]. \tag{7}$$

The exchange-correlation potential is then given by

$$V_{\rm xc}^{\rm tot}=\frac{\partial E_{\rm xc}^{\rm tot}}{\partial n}=\frac{\pi^2}{k_F^2}\frac{\partial E_{\rm xc}^{\rm tot}}{\partial k_F}. \tag{8}$$

Separating out the exchange part, which was worked out previously, we may express the correlation parts in the following form, where we have employed dimensionless units as in I, $\vec{k}=k_F\vec{x}$, $\vec{q}=k_F\vec{y}$, $\omega=k_Fuy$, $E_k=mE_x$, with $E_x=(1+\beta^2x^2)^{1/2}$, and $\beta=k_F/m$:

$$E_c^{\rm rings}=\frac{3Nk_F}{8\pi}\int_0^\infty y^3dy\int_{-\infty}^\infty du\left[\ln\left[1+\frac{e^2\beta}{\pi y^2}\tilde{Q}_{00}(y;u)\right]-\frac{e^2\beta}{\pi y^2}\tilde{Q}_{00}(y,u)\right]$$

$$+\frac{3Nk_F}{4\pi}\int_0^\infty y^3dy\int_{-\infty}^\infty du\left[\ln\left[1-\frac{2e^2\beta}{\pi y^2(1+u^2)}\tilde{Q}_T(y;u)\right]+\frac{2e^2\beta}{\pi y^2(1+u^2)}\tilde{Q}_T(y,u)\right] \tag{9}$$

and

$$V_c^{\rm rings}=\frac{e^2\beta k_F}{8\pi^2}\int_0^\infty y\,dy\int_{-\infty}^\infty du\tilde{Q}_{00}^{(1)}(y;u)\left[\frac{1}{1+(e^2\beta/\pi y^2)\tilde{Q}_{00}(y;u)}-1\right]$$

$$-\frac{e^2\beta k_F}{2\pi^2}\int_0^\infty y\,dy\int_{-\infty}^\infty\frac{du}{1+u^2}\tilde{Q}_T^{(1)}(y;u)\left[\frac{1}{1-[2e^2\beta/\pi y^2(1+u^2)]\tilde{Q}_T(y;u)}-1\right], \tag{10}$$

where

$$Q_{00}(q;\omega)=\frac{k_F^3}{4\pi^2m}\tilde{Q}_{00}(y;u)$$

and

$$Q_T(q;\omega)=\frac{k_F^3}{4\pi^2m}\tilde{Q}_T(y;u), \tag{11a}$$

$$\tilde{Q}_{00}(y;u)=\int_0^1 x^2dx\int_{-1}^{+1}d\mu\left[\frac{(E_{\vec{y}+\vec{x}}-E_x)[(E_{\vec{y}+\vec{x}}+E_x)^2-\beta^2y^2]}{E_xE_{\vec{x}+\vec{y}}[(E_{\vec{y}+\vec{x}}-E_x)^2+\beta^2u^2y^2]}\right.$$
$$\left.+\frac{(E_{\vec{y}+\vec{x}}+E_x)[(E_{\vec{y}+\vec{x}}-E_x)^2-\beta^2y^2]}{E_xE_{\vec{x}+\vec{y}}[(E_{\vec{y}+\vec{x}}+E_x)^2+\beta^2u^2y^2]}\right], \tag{11b}$$

$$\tilde{Q}_T(y;u)=\int_0^1 x^2dx\int_{-1}^{+1}d\mu\left[\frac{(E_{\vec{y}+\vec{x}}-E_x)[E_{\vec{x}+\vec{y}}E_x-\beta^2(\vec{x}\cdot\vec{y})^2/y^2-\beta^2(\vec{x}\cdot\vec{y})-1]}{E_{\vec{y}+\vec{x}}E_x[(E_{\vec{y}+\vec{x}}-E_x)^2+\beta^2u^2y^2]}\right.$$
$$\left.-\frac{(E_{\vec{y}+\vec{x}}+E_x)[E_{\vec{x}+\vec{y}}E_x+\beta^2(\vec{x}\cdot\vec{y})^2/y^2+\beta^2(\vec{x}\cdot\vec{y})+1]}{E_{\vec{y}+\vec{x}}E_x[(E_{\vec{y}+\vec{x}}+E_x)^2+\beta^2u^2y^2]}\right]. \tag{11c}$$

$\partial Q_{00}(q;\omega)/\partial k_F=k_F^2/4\pi^2m\tilde{Q}_{00}^{(1)}(y;u)$, where $\tilde{Q}_{00}^{(1)}(y;u)$ is just the μ integral of the integrand in (11b) and x set equal to 1, and $\partial Q_T(q;\omega)/\partial k_F=k_F^2/4\pi^2m\tilde{Q}_T^{(1)}(y;u)$, where $\tilde{Q}_T^{(1)}(y;u)$ is just the μ integral of the integrand in (11c) and x set equal to 1.

Several comments are in order at this stage.

(1) Expressions for $Q_{\lambda\mu}(q;\omega)$ have been derived in different ways by different authors quoted earlier who also express them in different forms. A little bit of algebra is required to show that they are all equivalent. The above form for the energy was given by Jancovici.[5]

(2) The longitudinal and transverse parts separate out neatly. In the nonrelativistic limit, $\beta<<1$, the transverse part is negligible being of order β^2 and the longitudinal part goes over to the expression derived by von Barth and Hedin[16] for the ring-sum contribution to the correlation energy.

(3) We have numerically integrated these expressions and they will be discussed in the next section. An important point to be mentioned here is that the numerical evaluation clearly exhibits exactly when the relativistic effects become comparable to the nonrelativistic calculation. This happens for $\beta\lesssim\frac{1}{4}$ for correlation energy and potential.

(4) Jancovici[5] has given an approximation for $\tilde{Q}_{00}$, $\tilde{Q}_T$ when $y<1$ and $\omega<E_F$, in the same spirit as Gell-Mann and Brueckner[3] did. The above expressions then simplify and analytic forms can be deduced. Jancovici made a further approximation of this and evaluated the results to order $e^4\ln e^2$.

The actual results without such an approximation are given here

$$\epsilon_c^{\text{rings}}\equiv\frac{E_c^{\text{rings}}}{N}\simeq\frac{3(k_F^2/m)}{32\pi(1+\beta^2)^{1/2}}\int_{-\infty}^{\infty}dv(\ln[1+R_L(v)]-R_L(v)-[R_L(v)]^2\{\ln[1+R_L(v)]-\ln R_L(v)\})$$
$$+\frac{3k_F}{16\pi}\int_{-\infty}^{\infty}dv(\ln[1+R_{\text{tr}}(v)]-R_{\text{tr}}(v)-[R_{\text{tr}}(v)]^2\{\ln[1+R_{\text{tr}}(v)]-\ln R_{\text{tr}}(v)\}), \tag{12}$$

where

$$R_L(v)=\frac{4e^2(1+\beta^2)^{1/2}}{\pi\beta}\left[1-v\tan^{-1}\frac{1}{v}\right], \tag{13a}$$

$$R_{\text{tr}}(v)=\frac{2e^2\beta}{\pi(1+\beta^2)^{1/2}(1+u^2)}\left[-u^2\frac{(\beta^2+1)}{\beta^2}+\frac{u(1+\beta^2)^{1/2}}{\beta}\left[1+\frac{u^2(\beta^2+1)}{\beta^2}\right]\tan^{-1}\left[\frac{\beta}{u(\beta^2+1)^{1/2}}\right]\right], \tag{13b}$$

and

$$V_c^{\text{rings}}=\epsilon_c^{\text{rings}}+\frac{\beta}{3}\frac{\partial}{\partial\beta}\epsilon_c^{\text{rings}} . \quad (14)$$

These integrals are evaluated numerically. For $\beta << 1$, they reduce to the correct nonrelativistic expressions and $\epsilon_c^{\text{rings}} \simeq \epsilon_c$(GB). Jancovici gave expressions to order $e^4 \ln e^2$ which are obtained by just keeping only the corresponding terms in Eq. (12), and thus missed a density-dependent term. The purpose of the above derivation is to show that an appproximation in the spirit of GB leads to the correct nonrelativistic answer for $\beta << 1$ as well as contains Jancovici's result. The actual numerical answers obtained in this way are displayed in Fig. 1 with the Coulomb and transverse-photon contributions shown separately for the correlation energy where the full-line curve represents Eq. (9), long-dashed curve, Eq. (12), and short-dashed curve represents the $e^4 \ln e^2$ approximation. For comparison, we have also given the nonrelativistic von-Barth—Hedin curve. In Fig. 2, only our calculations are given for the correlation potential, Eq. (10), and Jancovici's approximations lie in a relation similar to the correlation-energy results of Fig. 1. It should be pointed out that the formal expressions given above are in relativistic units

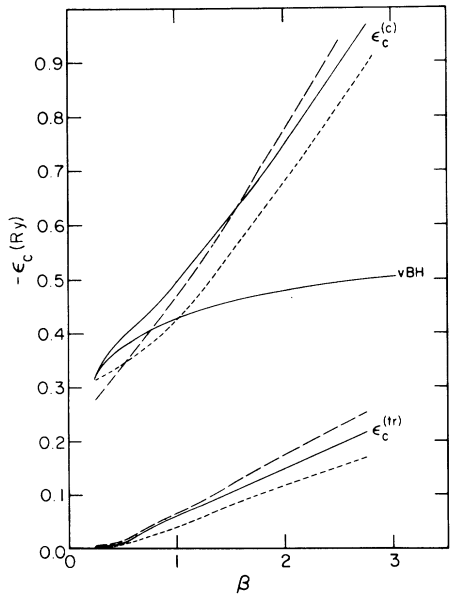

FIG. 1. The correlation energy expressed in rydbergs versus β ($=\hbar k_F/mc$). $\epsilon_c^{(C)}$ stands for the Coulomb, $\epsilon_c^{(tr)}$ for the transverse-photon contributions. The full-line curves are our results for $\epsilon_c^{(C)}$ and $\epsilon_c^{(tr)}$; the long-dashed curves are the result of Eq. (12) and the short-dashed curves are the the $e^4 \ln e^2$ approximation of Jancovici. The nonrelativistic answer is labeled vBH.

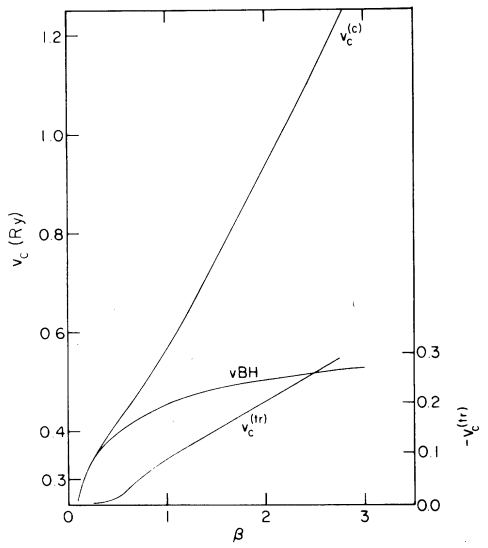

FIG. 2. The correlation potential in rydbergs versus β. $V_c^{(C)}$, $V_c^{(tr)}$, and vBH stand for Coulomb, transverse-photon, and von-Barth—Hedin results. The scale on the right should be used for $V_c^{(tr)}$.

($\hbar=1=c$ and $a_0=$ Bohr radius $=1$), and a Rydberg is $\frac{1}{2}e^2$ ($e^2=1/137.037$) relativistic units of energy.

III. DISCUSSION OF THE RESULTS

In Fig. 1 we have plotted the correlation energy contributions as a function of β, when the ring sums are computed numerically (full-line curves) and when an approximation due to Jancovici [Eq. (12)] is used (long-dashed curves). Also given is the nonrelativistic result evaluated from the expression given by von Barth and Hedin[16] (vBH) which corresponds to the nonrelativistic limit of our expressions. Also given is the result to order $e^4 \ln e^2$ (short-dashed lines) obtained by Jancovici. The Jancovici schemes are in the same spirit as the Gell-Mann—Brueckner[3] (GB) one involving a small-q approximation for the Q_0, Q_T but retaining the ω dependence. The region of our interest (heavy-atom systems) corresponds to $0.25 < \beta < 2.75$.

(i) In the nonrelativistic case, we have verified that the GB approximation is poor (20—30% of the vBH answers) and so the q dependence is important. On the other hand, the long-dashed curve lies 8% above our full-line curve at $\beta=2.75$ for $\epsilon_c^{(C)}$, crosses it at about $\beta=1.6$, and lies 15% below for $\beta=0.25$; but the long-dashed curve lies above our $\epsilon_c^{(tr)}$ in the entire β range, being 17% at $\beta=2.75$. The $e^4 \ln e^2$ approximation, on the other

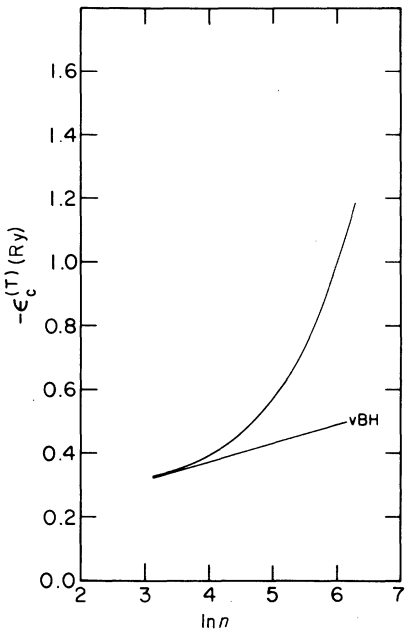

FIG. 3. The total correlation energy in rydbergs as a function of ln$n$, where n = density $\times a_0^3$, a_0 being the Bohr radius. vBH is the corresponding nonrelativistic result.

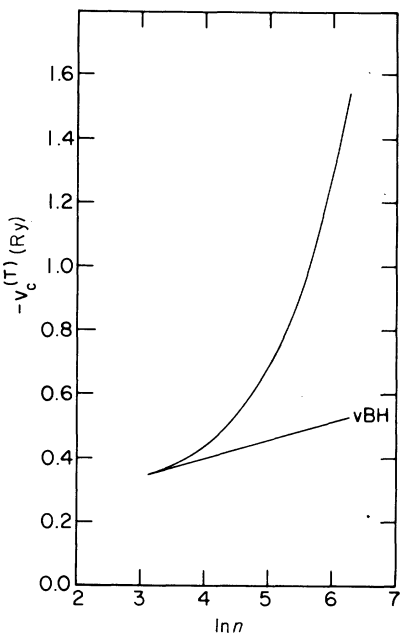

FIG. 4. The total correlation potential versus ln$n$; vBH is the corresponding nonrelativistic approximation.

hand, lies below our curve for both $\epsilon_c^{(C)}$ (8% for $\beta=2.75$ and 3% for $\beta=0.25$) and $\epsilon_c^{(\mathrm{tr})}$ (21% for $\beta=2.75$) in the entire β range. Thus, for the individual $\epsilon_c^{(C)}$ and $\epsilon_c^{(\mathrm{tr})}$ as well as for the total ϵ_c, the q dependence does not appear to be as bad an approximation as with the nonrelativistic case, being of order 8% in the Jancovici scheme and 10% in the $e^4\ln e^2$ scheme from ours. Similar remarks apply to the potential.

(ii) The nonrelativistic result is very different from the relativistic for $\beta > 0.25$, as expected. In Fig. 3, the total correlation energy per particle versus ln$n$ is plotted, where n is the electron density in atomic units (density multiplied by a_0^3). The correlation potential as a function of β is given in Fig. 2, where the individual Coulomb and

TABLE I. The values of correlation energy and potential in rydbergs are given for a set of β values of interest to heavy atom systems. $\epsilon_C^{(T)}=\epsilon_c^{(C)}+\epsilon_c^{(\mathrm{tr})}$ and $V_c^{(T)}=V_c^{(C)}+V_c^{(\mathrm{tr})}$. The superscript ($C$) and superscript (tr) stand for the Coulomb and transverse-photon contributions. For comparison we have also given the von-Barth–Hedin (vBH) results.

β	$-\epsilon_c^{(\mathrm{vBH})}$	$-\epsilon_c^{(C)}$	$-\epsilon_c^{(\mathrm{tr})}$	$-\epsilon_c^{(T)}$	$-v_c^{(\mathrm{vBH})}$	$-v_c^{(C)}$	$-v_c^{(\mathrm{tr})}$	$-v_c^{(T)}$
0.007	0.1234	0.1235		0.1235	0.140	0.1389		0.1389
0.014	0.1573	0.1574		0.1574	0.173	0.174		0.174
0.25	0.3252	0.3262	0.0036	0.3298	0.347	0.3453	0.0064	0.3517
0.50	0.3739	0.3924	0.0075	0.3999	0.399	0.4209	0.0131	0.4340
0.75	0.4046	0.4370	0.0369	0.4739	0.430	0.4918	0.0586	0.5504
1.0	0.4269	0.4997	0.0595	0.5592	0.453	0.5721	0.0967	0.6688
1.25	0.4442	0.5546	0.0828	0.6374	0.470	0.6581	0.1218	0.7799
1.5	0.4581	0.6188	0.1062	0.7250	0.483	0.7489	0.1530	0.9019
1.75	0.4697	0.6857	0.1292	0.8149	0.494	0.8336	0.1825	1.0161
2.0	0.4794	0.7545	0.1519	0.9164	0.503	0.9373	0.2119	1.1492
2.25	0.4878	0.8249	0.1740	0.9989	0.511	1.0356	0.2409	1.2765
2.5	0.4951	0.8965	0.1964	1.0929	0.518	1.1339	0.2695	1.4034
2.75	0.5015	0.9690	0.2182	1.1872	0.523	1.2329	0.2979	1.5308

transverse contributions are separately shown as well as the von-Barth—Hedin answer for comparison. In Fig. 4, the total correlation potential as a function of $\ln n$ is given. The departure from the nonrelativistic result begins at $\beta > 0.25$ as in the energy case. It should be pointed out for both energy and potential, the transverse-photon contribution though small, is found to be significant.

(iii) In Table I, we have given the numerical values for energy per particles and the potential for some values of β along with the results of von Barth and Hedin. The separate contributions of the Coulomb and transverse photons are also given for completeness . In I, it was found that in the case of exchange, the two contributions are comparable to each other and are of opposite sign. In contrast to this, the correlation contributions are of the same negative sign. By a comparison of the orders of magnitude, we may infer that the total correlation contributions to the potential are about 5% of the total exchange for $\beta = 2.5$ and 2.5% for $\beta = 1$.

(iv) In the past, Freedman *et al.*[17] and Fricke *et al.*[18] have used the nonrelativistic GB result to estimate the correlation effects in fermium to be less than 1 eV. The present work while showing that the GB result is small in comparison to the actual relativistic result, leads one to believe that it is perhaps of the order of 10 eV. Our preliminary calculations for fm show that the 1*s* binding energy is lowered by about 6 eV when correlations are included.

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