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Gradient expansion in the density functional approach to an inhomogeneous electron system

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The linearized integral equation obeyed by the irreducible vertex function associated with the density fluctuations is solved *exactly* up to second power in the wave vector. This is used to compute the static polarizability of the homogeneous electron system up to this order. It determines the gradient expansion coefficient in the density-functional formalism for the inhomogeneous electron system. The result is compared with existing approximate caclulations. The method of solution is applicable to irreducible vertex functions which appear in the determination of other correlation functions of the homogeneous systems. Our gradient term vanishes for both extreme high- and low-density regions, unlike the results of Kleinman and Sham.

I. INTRODUCTION

Almost a decade ago, one of us[1] developed methods of investigating the integral equation obeyed by an irreducible vertex function $\Gamma_{\uparrow,\downarrow}(k;q)$ associated with the transverse spin susceptibility of an itinerant magnetic electron system. In the long-wavelength limit $q \to 0$, it was found that one may expand this vertex function in a power series in the wave vector, the coefficients of each power of which is a spherical harmonic expansion. In this way, the q^2 coefficient of the spin-wave dispersion is found to be an integral over $\Gamma_{10}^{(1)}(k)$, where the superscript indicates the coefficients of q in the $\Gamma_{\uparrow,\downarrow}(k;q)$ expansion and the subscripts 10 mean the p-wave part with zero projection in the spherical harmonic expansion. It was then found that $\Gamma_{10}^{(1)}(k)$ obeys a linear integral equation which in turn was solved by a variational method, since a series solution in k of this equation was not found satisfactory. In this manner, the effects of interactions (including all renormalizations) among the electrons are taken into account almost completely within the scheme of the linearized-vertex-function formalism. To investigate the spin-density-wave (SDW) instability of this system, the solution of the static part of this same vertex equation for finite wave vectors is required. To examine this, we developed a variational method for $\Gamma_{\uparrow,\downarrow}(k;q)$ directly and deduced from this a simple criterion for the SDW instability. The latter method was found very powerful in dealing with other vertex functions, and the author with his collaborators has examined other properties of the interacting electron system,[2,3] as well as its relationship with other methods such as moment-conserving schemes.[4,5]

In the density-functional approach to the inhomogeneous electron systems,[6-9] one finds that the better the knowledge of various correlation functions,[9] the better the description of the inhomogeneous system. For instance, in Ref. 9, we used the Yukawa interaction model in the limit of large screening to deduce the spin splitting of energy bands in ferromagnets. In the paramagnetic case, in particular, Sham[10] has given a calculation of the static density-correlation function up to q^2 terms by solving the integral equation obeyed by the associated irreducible vertex function by one iteration. The purpose of this paper is to use the methods of Ref. 1 to determine the same quantity that Sham computed both by an exact solution of the vertex equation and by the variational method for which the general expression for all q for the quantity of interest already exists.[3] In contrast to the spin-wave calculation, *it is found here that the vertex equation can be solved exactly in this limit.* All the integrals appearing in the theory can be done generally for a wide class of static interaction potentials. The detailed calculation for the Yukawa potential is derived from the general expressions. This therefore serves to determine how good the simple variational answer is to this order, as well as derive an exact expression for the coefficient of the square of the gradient of the density in the theory of the inhomogeneous electron system. Kleinman[11] has very recently developed a solution to the vertex equation to this order. He has criticized Sham's[10] work in the light of the results he has derived. We will here make a critical assessment of the situation.

In Sec. II, we give only a brief description of the solution to the vertex equation appropriate to the problem at hand. Elsewhere[12] we will give a complete account of this method and its application to other properties of the system. In Sec. III, the variational solution[3] is given to this order. The results are compared with each other in Sec. IV. In Sec. V, we specialize the results for the Yukawa interaction model, and we give a critical assessment of these expressions in relation to the works

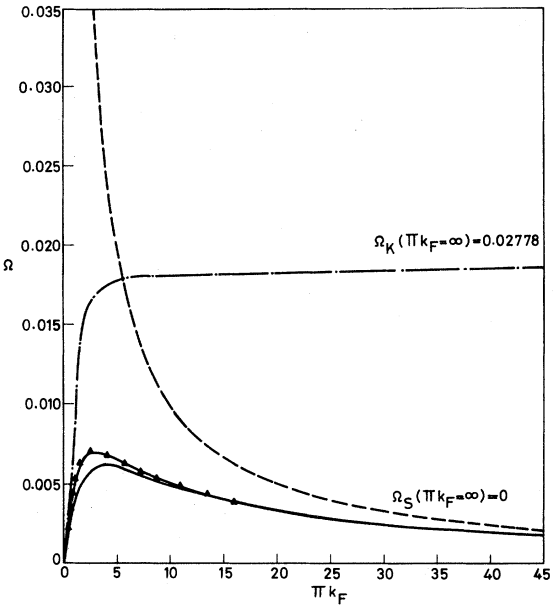

FIG. 1. Graph of Ω as a function of πk_F in Hartree units; Sham's: dashed curve; Kleinman's: dot-dashed curve; exact, Thomas-Fermi: solid-triangle curve; exact, self-consistent: solid-line curve.

of Sham[10] and Kleinman.[11] In this section, we also point out the adequacy of the schemes for calculating the screening parameter in the Yukawa model.[13] This is because with a pure Coulomb potential for the electron interactions, there is a divergence characteristic of the long-range nature of this potential.[10,11] A standard way to handle this is to introduce a screened Coulomb potential instead; if the screening is treated as a density-independent parameter, in the limit of it becoming zero, this divergence persists as Kleinman has shown, which is also borne out by our analysis. The forms of the screening as a function of density, however, can be treated in some schemes which are discussed in Sec. V. The quantity of interest in the gradient expansion is Ω (in the notation of Sham[10]) as a function of density, and it is given in Fig. 1. For comparison, we have also plotted on the same graph those given by Sham and Kleinman. As shown in this paper, Sham's analysis is exact for high densities and is zero in this limit; we are in agreement with this result in contrast to Kleinman. For low densities, Kleinman's analysis is correct within the density-dependent screening model where Ω again approaches zero. Sham's procedure is inapplicable here, as his analysis was specially designed for high densities, as shown in Sec. V. These features are brought out explicitly in our analysis. Section V also contains details of these calculations. These various features are due to a proper combination of the renormalizations *and* to the screening being a function of density in the form specified in Sec. V. Even more important, the antiparallel spin correlations are not taken into account in a linearized vertex equation, and only recently have we made progress in a variational solution to the nonlinear vertex equation.[14] We also present here a short summary of the results obtained.

II. SOLUTION OF THE IRREDUCIBLE VERTEX EQUATION

The longitudinal static dielectric function may be expressed in terms of the irreducible density-correlation function and the associated irreducible vertex function as

$$\epsilon_L(q) = 1 + (4\pi/q^2)\chi(q), \tag{1}$$

where

$$\chi(q) = 2e^2 \int \mathcal{F}(k, q)\Gamma(k;q)[d^3k/(2\pi)^3]. \tag{2}$$

Here $\mathcal{F}(k, q)$ is given by

$$\mathcal{F}(k, q) = -\frac{f_0(\vec{k}+\frac{1}{2}\vec{q}) - f_0(\vec{k}-\frac{1}{2}\vec{q})}{\epsilon_{\vec{k}+1/2\vec{q}} - \epsilon_{\vec{k}-1/2\vec{q}}}, \tag{3}$$

with ϵ_k the noninteracting one-electron energy, and $f_0(k)$ the Fermi function associated with the interacting electron of state $\vec{k}$. $\Gamma(k;q)$ obeys the linear integral equation in the random-phase approximation when the antiparallel spin correlations are neglected,[14]

$$\Gamma(k;q) = 1 + \int v_s(|\vec{k}-\vec{k}_1|)\mathcal{F}(k_1, q) \times \left[\Gamma(k_1;q) - \left(\frac{\epsilon_{\vec{k}_1+\vec{q}/2} - \epsilon_{\vec{k}_1-\vec{q}/2}}{\epsilon_{\vec{k}+\vec{q}/2} - \epsilon_{\vec{k}-\vec{q}/2}}\right)\Gamma(k;q)\right]\frac{d^3k_1}{(2\pi)^3} \tag{4}$$

This equation is the same one that Sham[10] and Kleinman[11] considered except that we have written it in such a way as to display in an explicit way the correlation contribution to the one-electron energy. It is for this reason we use the notation Γ instead of their $\tilde{\Lambda}$. The advantages of doing this will become evident shortly. The interaction potential $v_s(|\vec{k}-\vec{k}_1|)$ represents the statically screened electron-electron interaction. It is for this reason $\Gamma(k;q)$ does not depend on the frequency part associated with k. We shall not specify its form here.

We will now express these equations in dimensionless form, as is done in Ref. 1. Thus, in the notation of Ref. 1, we have

$$\chi(q) = \left(\frac{me^2k_F}{2\pi^2}\right)\int x^{1/2}\,dx \int \frac{d\hat{x}}{4\pi}\mathcal{F}(x, y)\Gamma(x; y), \tag{5}$$

and

$$\Gamma(x; y) = 1 + \left(\frac{2\alpha r_s}{\pi}\right)\int x_1^{1/2}\,dx_1 \int \frac{d\hat{x}_1}{4\pi} V_s(|\vec{x}-\vec{x}_1|)$$

$$\times \mathcal{F}(x_1; y)\left[\Gamma(x_1; y) - \left(\frac{x_1}{x}\right)^{1/2} \frac{\cos\theta_{x_1}}{\cos\theta_x}\Gamma(x; y)\right]. \tag{6}$$

Here

$$v_s(|\vec{k} - \vec{k}_1|) = \frac{4\pi e^2}{k_F^2} V_s(|\vec{x} - \vec{x}_1|)$$

$$= \frac{4\pi e^2}{k_F^2} \sum_{l,m} V_s^{(l)}(x; x_1) Y_{lm}(\hat{x}) Y_{lm}^*(\hat{x}_1), \tag{7}$$

where we have put $|\vec{x}|^2 = x$, etc. Here r_s is the usual dimensionless electron-gas parameter related to k_F via $\alpha r_s a_0 k_F = 1$, a_0 being the Bohr radius, $\alpha = (4/9\pi)^{1/3}$.

We would like to draw attention to one necessary approximation that seems to be made by all without much comment except for Kleinman,[11] who seems to make the approximation too when it comes to the actual evaluation of integrals. The expression for the one-electron energy is itself a complicated nonlinear integral equation of the Hammerstein type, even in the Hartree-Fock (HF) approximation. Recall that in the HF scheme, the one-electron energy has for the interaction contribution, an integral over the interaction potential in momentum space over an energy surface specified by the *exact* HF energy due to the appearance of the Fermi function $f_0(k)$. We have shown elsewhere[14] that, to a *very good approximation*, one may take this $f_0(k)$ to be the one for the free-electron states; in fact, this approximation is a variational one, but the energy renormalization is taken care of in the following way. It is the second term in square brackets in Eq. (6), which takes into account the one-electron energy renormalization, an important feature which is responsible for making all the integrals devoid of divergences in the subsequent analysis. This feature of the rewriting of the irreducible vertex equation has therefore incorporated the single-particle energy renormalization appropriately. (It may be noted that the difference of the actual one-particle energy and Fermi energy in the HF scheme for the Coulomb gas, for instance, *can be shown* to be a product of a positive function of the wave vector times the difference in the wave vector and the Fermi wave vector. In spite of this, one cannot hope to get an exact solution of Eq. (6) without solving the one-electron energy exactly also, which is impossible. But for making this important observation, we shall not dwell on this point again in subsequent discussions.)

In the long-wavelength limit, to order q^2, we have (see, for instance, Ref. 2) ($q \equiv y k_F$),

$$\mathcal{F}(x, y) \cong \delta(x-1) + \tfrac{1}{4}y^2[\delta'(x-1) + \tfrac{2}{3}x\cos^2\theta_x \delta''(x-1)], \tag{8}$$

where the primes denote differentiation with respect to x. We now write to this order (in view of spherical and inversion symmetries in the homogeneous system),

$$\Gamma(x; y) = \sum_{l,m}[\Gamma_{lm}^{(0)}(x) + y^2\Gamma_{lm}^{(2)}(x)]Y_{lm}(\hat{x}). \tag{9}$$

Using the standard formulas concerning spherical harmonics, etc., putting these in Eq. (6), and equating like powers of y on both sides of the resulting equation, we obtain the ones for $\Gamma_{lm}^{(l)}(x)$; that is

$$\Gamma_{lm}^{(0)}(x) = (4\pi)^{1/2}\delta_{l,0}\delta_{m,0} + \left(\frac{2\alpha r_s}{\pi}\right)\left(\Gamma_{lm}^{(0)}(1)\frac{V_s^{(l)}(x;1)}{2l+1} - \frac{1}{x^{1/2}}\frac{V_s^{(1)}(x;1)}{3}\Gamma_{lm}^{(0)}(x)\right). \tag{10}$$

This can be solved at once. We introduce the following abbreviations:

$$\Gamma_{lm}^{(0)}(x) = (4\pi)^{1/2}\Gamma\tilde{\Gamma}(x)\delta_{l,0}\delta_{m,0}, \tag{11}$$

where

$$\Gamma = 1/\{1 - (2\alpha r_s/\pi)[V_s^{(0)}(1;1) - \tfrac{1}{3}V_s^{(1)}(1;1)]\}, \tag{12}$$

and

$$\tilde{\Gamma}(x) = \frac{1 - (2\alpha r_s/\pi)[V_s^{(0)}(1;1) - V_s^{(0)}(x;1)] + (2\alpha r_s/\pi)\tfrac{1}{3}V_s^{(1)}(1;1)}{1 + (2\alpha r_s/\pi)x^{-1/2}\tfrac{1}{3}V_s^{(1)}(x;1)}. \tag{13}$$

Observe that $\tilde{\Gamma}(x=1) = 1$. This solution is obtained, provided that

$$1 \neq \frac{2\alpha r_s}{\pi}\left(\frac{V_s^{(l)}(1;1)}{2l+1} - \frac{V_s^{(1)}(1;1)}{3}\right). \tag{14}$$

Γ is recognized to be the vertex renormalization constant at the Fermi surface. $\Gamma_{lm}^{(0)}(x)$ is thus zero for all but $l=0$, $m=0$, a result which helps us enormously in the subsequent analysis.

Similarly, we obtain, in view of Eq. (11), the equation obeyed by $\Gamma_{lm}^{(2)}(x)$ also as an algebraic equation,

$$\Gamma_{lm}^{(2)}(x) = \left(\frac{2\alpha r_s}{\pi}\right)\left(\Gamma_{lm}^{(2)}(1)\frac{V_s^{(l)}(x;1)}{2l+1} - x^{-1/2}\frac{V_s^{(1)}(x;1)}{3}\Gamma_{lm}^{(2)}(x)\right)$$

$$+ \frac{2\alpha r_s}{\pi}\left(-\frac{1}{4}\frac{d}{dx_1}\left\{x_1^{1/2}\left[\Gamma_{00}^{(0)}(x_1)V_s^{(0)}(x;x_1) - \left(\frac{x_1}{x}\right)^{1/2}\Gamma_{00}^{(0)}(x)\frac{V_s^{(1)}(x;x_1)}{3}\right]\right\}\right.$$

$$+\frac{1}{18}\frac{d^2}{dx_1^2}\left\{x_1^{3/2}\left[\Gamma_{00}^{(0)}(x_1)V_s^{(0)}(x;x_1)+\frac{4}{5}\left(\frac{x_1}{x}\right)^{1/2}\Gamma_{00}^{(0)}(x)\frac{V_s^{(3)}(x;x_1)}{7}-\frac{9}{5}\left(\frac{x_1}{x}\right)^{1/2}\Gamma_{00}^{(0)}(x)\frac{V_s^{(1)}(x;x_1)}{3}\right]\right\}\Bigg)_{x_1=1}\delta_{l,0}\delta_{m,0}$$

$$+\frac{2\alpha r_s}{\pi}\left(\frac{1}{6}\frac{d^2}{dx_1^2}\left\{x_1^{3/2}\left[\frac{1}{3}\Gamma_{00}^{(0)}(x_1)\left(\frac{4}{5}\right)^{1/2}\frac{V_s^{(2)}(x;x_1)}{5}-\frac{1}{3}\left(\frac{x_1}{x}\right)^{1/2}\Gamma_{00}^{(0)}(x)\left(\frac{4}{5}\right)^{1/2}\frac{V_s^{(3)}(x;x_1)}{7}\right]\right\}\right)_{x_1=1}\delta_{l,2}\delta_{m,0}. \quad (15)$$

Once again, provided that Eq. (14) holds, we obtain an explicit solution for $\Gamma_{lm}^{(2)}(x)$ *which is nonzero only for* $l=0,2$ and $m=0$.

We now obtain for $\chi(q)$ the expression calculated to second order in q,

$$\chi(q)\cong\left(\frac{me^2k_F}{4\pi^3}\right)(4\pi)^{1/2}\left\{\Gamma_{00}^{(0)}(1)+y^2\left[\Gamma_{00}^{(2)}(1)-\frac{1}{4}\frac{d}{dx}[x^{1/2}\Gamma_{00}^{(0)}(x)]+\frac{1}{18}\frac{d^2}{dx^2}[x^{3/2}\Gamma_{00}^{(0)}(x)]+\frac{1}{18}\left(\frac{4}{5}\right)^{1/2}\frac{d^2}{dx^2}[x^{3/2}\Gamma_{20}^{(0)}(x)]\right]_{x=1}\right\}.$$

In view of the solutions obtained above, we finally express in the form

$$\chi(q)\cong\left(\frac{me^2k_F}{\pi^2}\right)\Gamma\left(1+\frac{y^2}{4}\left\{\left(-\frac{d}{dx}[x^{1/2}\tilde{\Gamma}(x)]+\frac{2}{9}\frac{d^2}{dx^2}[x^{3/2}\tilde{\Gamma}(x)]\right)_{x=1}+\left(\frac{2\alpha r_s}{\pi}\right)\Gamma\left[-\frac{d}{dx}\left(x^{1/2}\tilde{\Gamma}(x)V_s^{(0)}(x;1)\right.\right.\right.$$

$$\left.\left.\left.-x\frac{V_s^{(1)}(x;1)}{3}\right)+\frac{2}{9}\frac{d^2}{dx^2}\left(x^{3/2}\tilde{\Gamma}(x)V_s^{(0)}(x;1)+\frac{4}{5}x^2\frac{V_s^{(3)}(x;1)}{7}-\frac{9}{5}x^2\frac{V_s^{(1)}(x;1)}{3}\right)\right]_{x=1}\right\}\right). \quad (16)$$

This is the result of an exact solution to the irreducible equation, Eq. (6). In Sec. III, we will merely give the variational result calculated to the same order, and compare it with Eq. (16).

III. VARIATIONAL RESULT AND COMPARISON WITH OTHER RESULTS

In Ref. 3, the vertex equation, Eq. (6), is solved by a variational method, and $\chi(q)$ is computed for general wave vector q, frequency ω. In this, we set $\omega=0$ and compute the static expression up to q^2 order. Without giving the details of computation, we quote the final result here, in the present notation,

$$\chi_{\rm var}(q)\cong\left(\frac{me^2k_F}{\pi^2}\right)\Gamma\left(1+\frac{y^2}{4}\left\{-\frac{2}{3}+\frac{1}{3}\Gamma+\left(\frac{2\alpha r_s}{\pi}\right)\Gamma\left[-\frac{d}{dx}\left(2x^{1/2}V_s^{(0)}(x;1)-(x+1)\frac{V_s^{(1)}(x;1)}{3}\right)\right.\right.$$

$$\left.\left.+\frac{2}{9}\frac{d^2}{dx^2}\left(2x^{3/2}V_s^{(0)}(x;1)-\frac{9}{5}x^2\frac{V_s^{(1)}(x;1)}{3}-x\frac{V_s^{(1)}(x;1)}{3}+\frac{4}{5}x^2\frac{V_s^{(3)}(x;1)}{7}\right)\right]_{x=1}\right\}\right), \quad (17)$$

It may be of interest to point out that the variational solution to the vertex equation has the structure

$$\Gamma_{\rm var}(x;y)=I(y)/[I(y)-J(y)], \quad (18)$$

where

$$I(y)=\int x^{1/2}\,dx\int d\hat{x}\,\mathcal{F}(x;y) \quad (18a)$$

and

$$J(y)=(\alpha r_s/2\pi^2)\int x^{1/2}\,dx\int d\hat{x}\int x_1^{1/2}\,dx_1\int d\hat{x}_1$$

$$\times\mathcal{F}(x,y)\mathcal{F}(x_1,y)V_s(|\vec{x}-\vec{x}_1|)$$

$$\times[1-(x_1/x)^{1/2}\cos\theta_{x_1}/\cos\theta_x]. \quad (18b)$$

Observe that the trial solution $\Gamma_{\rm var}(x;y)$ has no x dependence at all. When we compare Eqs. (16) and (17), we will be surprised to find that the difference between them has a simple form:

$$\chi(q)=\chi_{\rm var}(q)+y^2\left(\frac{me^2k_F}{\pi^2}\right)\frac{1}{9}\Gamma^2\left(\frac{2\alpha r_s}{\pi}\right)^2 Z_1$$

$$\times\left[\frac{d}{dx}\left(V_s^{(0)}(x;1)-x^{-1/2}\frac{V_s^{(1)}(x;1)}{3}\right)\right]_{x=1}^2. \quad (19)$$

Here

$$Z_1=1/[1+(2\alpha r_s/\pi)\tfrac{1}{3}V_s^{(1)}(1;1)]. \quad (20)$$

In deriving Eq. (19), one makes use of many simplifying properties that $\tilde{\Gamma}(x)$ enjoys, even though the actual computation is somewhat tedious.

In Sec. IV, we will discuss in detail the various aspects of a very popular model for $V_s(|\vec{x}-\vec{x}_1|)$, namely the Yukawa potential.

IV. COMPUTATION OF $g_{\rm xc}^{(2)}$ OF THE GRADIENT EXPANSION

Sham[10] has shown that if one writes $\chi(q)$ in the form (we use the notations as in Sham's paper)

$$\chi(q)\cong\chi^{(0)}+q^2\chi^{(2)}, \quad (21)$$

then the coefficient $g_{\rm xc}^{(2)}(n)$ of the gradient expansion in the theory of the inhomogeneous electron system is

$$g_{\rm xc}^{(2)}(n)=[\chi^{(2)}/(\chi^{(0)})^2-\chi_0^{(2)}/(\chi_0^{(0)})^2], \quad (22)$$

where the subscripts 0 indicate the same quantities for the noninteracting system with the same density.

If $E_{xc}[n]$ is the exchange and correlation energy functional, Kleinman defined[11]

$$E_{xc}[n]=\int \epsilon_{xc}(n) n\, d^3r+\tfrac{1}{2}\int g_{xc}^{(2)}(n)\,|\vec{\nabla} n|^2\, d^3r+\cdots,$$

where $\epsilon_{xc}(n)$ is the exchange and correlation energy per electron in the uniform gas. This serves to define $g_{xc}^{(2)}$. The latter are easily calculated because in the expressions for $\chi^{(0)}$, $\chi^{(2)}$, if we set the terms containing $V_s^{(1)}$ zero, we obtain $\chi_0^{(0)}$, $\chi_0^{(2)}$, respectively.

We first make the observation that if we employ the first iterative solution to the $\Gamma(x;y)$ equation as was done by Sham,[10] we obtain

$$\Gamma(x;y)\cong 1+\left(\frac{\alpha r_s}{2\pi^2}\right)\int x_1^{1/2}\,dx_1\int d\hat{x}_1 V_s(|\vec{x}-\vec{x}_1|)\times \mathfrak{F}(x_1;y)\left[1-\left(\frac{x_1}{x}\right)^{1/2}\frac{\cos\theta_{x_1}}{\cos\theta_x}\right]. \tag{23}$$

Using this in Eq. (5) and the notations (18a, 18b), we obtain

$$\chi_{\mathrm{Sham}}(q)\cong (mk_F e^2/4\pi^3)[I(y)+J(y)]. \tag{24}$$

Even though the variational solution is of the form (18), in contrast to the iterative solution (23), we observe that $\chi_{\mathrm{var}}(q)$ is found to be

$$\chi_{\mathrm{var}}(q)=\left(\frac{mk_F e^2}{4\pi^3}\right)\frac{[I(y)]^2}{I(y)-J(y)}. \tag{25}$$

Sham's procedure is equivalent to expanding $\chi(q)$ up to first order in $\alpha r_s/\pi$, and so expanding $\chi_{\mathrm{var}}(q)$ up to this order. We observe this to be the *same expression as Sham's*.

On the other hand, if we use $\chi_{\mathrm{var}}(q)$ to compute it to order q^2, we obtain

$$\chi_{\mathrm{var}}(q)\cong\left(\frac{mk_F e^2}{4\pi^3}\right)\Gamma I^{(0)}\left\{1+\left[2\frac{I^{(2)}}{I^{(0)}}-\Gamma\left(\frac{I^{(2)}}{I^{(0)}}-\frac{J^{(2)}}{I^{(0)}}\right)\right]y^2\right\}, \tag{26}$$

and Sham obtains

$$\chi_{\mathrm{Sham}}(q)\cong\left(\frac{mk_F e^2}{4\pi^3}\right)I^{(0)}\left[1+\frac{J^{(0)}}{I^{(0)}}+\left(\frac{I^{(2)}}{I^{(0)}}+\frac{J^{(2)}}{I^{(0)}}\right)y^2\right]. \tag{27}$$

This essentially means that the vertex renormalization $\Gamma\,[\equiv(1-J^{(0)}/I^{(0)})^{-1}]$ is set equal to $1+J^{(0)}/I^{(0)}$ in Sham's procedure. Since setting $J=0$ and $\Gamma=1$ imply that we are dealing with the noninteracting system, we obtain for $g_{xc}^{(2)}$ the following expressions (in our notation):

$$g_{xc}^{(2)}(\mathrm{var})=4\pi^3/mk_F e^2 I^{(0)}\Gamma\times\{2(I^{(2)}/I^{(0)})[(1-\Gamma)/\Gamma]+J^{(2)}/I^{(0)}\}, \tag{28a}$$

and

$$g_{xc}^{(2)}(\mathrm{exact})=\frac{4\pi^3}{mk_F e^2 I^{(0)}\Gamma}\left\{2\frac{I^{(2)}}{I^{(0)}}\left(\frac{1-\Gamma}{\Gamma}\right)+\frac{J^{(2)}}{I^{(0)}}+\left(\frac{2\alpha r_s}{\pi}\right)^2 Z_1\left[\frac{d}{dx}\left(V_s^{(0)}(1;x)-x^{-1/2}\frac{V_s^{(1)}(1;x)}{3}\right)\right]_{x=1}^2\right\}$$
$$\equiv g_{xc}^{(2)}(\mathrm{var})+\frac{4\pi^3}{mk_F e^2 I^{(0)}\Gamma}\left(\frac{2\alpha r_s}{\pi}\right)^2 Z_1\left[\frac{d}{dx}\left(V_s^{(0)}(1;x)-x^{-1/2}\frac{V_s^{(1)}(1;x)}{3}\right)\right]_{x=1}^2. \tag{28b}$$

Now from Eq. (17), we may deduce

$$I^{(0)}=4\pi,\quad I^{(2)}/I^{(0)}=-\tfrac{1}{12},$$
$$\Gamma=\left[1-\left(\frac{2\alpha r_s}{\pi}\right)[V_s^{(0)}(1;1)-\tfrac{1}{3}V_s^{(1)}(1;1)]\right]^{-1},$$

and

$$\frac{J^{(2)}}{I^{(0)}}=\left(\frac{2\alpha r_s}{\pi}\right)\left[-\frac{1}{4}\frac{d}{dx}\left(2x^{1/2}V_s^{(0)}(1;x)-(1+x)\frac{V_s^{(1)}(1;x)}{3}\right)+\frac{1}{18}\frac{d^2}{dx^2}\left(2x^{3/2}V_s^{(0)}(1;x)-\frac{9}{5}x^2\frac{V_s^{(1)}(1;x)}{3}+\frac{4}{5}x^2\frac{V_s^{(3)}(1;x)}{7}-x\frac{V_s^{(1)}(1;x)}{3}\right)\right]_{x=1}. \tag{29}$$

Thus we see that the variational result is obtained when Z_1 is set equal to zero, and Sham's answer is obtained when Z_1 is set equal to zero, and Γ equal to $1+J^{(0)}/I^{(0)}$, and working to first power in $\alpha r_s/\pi$ (high-density limit) in the exact expressions. The correction to the variational answer is thus seen to be of order $(\alpha r_s/\pi)^2$ including powers of $\alpha r_s/\pi$ in Eq. (28b) without those appearing in Γ and Z_1, as they are of magnitude unity plus corrections of order $\alpha r_s/\pi$ provided that the expressions multiplying them are finite in this limit. In Sec. V, we will specialize the above results in terms of a Yukawa form for $V_s(|\vec{k}-\vec{k}_1|)$ and discuss the expressions in various regions of validity of such a model. We may state here that by expressing our results in terms of $V_s^{(1)}$ as above, we

have made a contact with Fermi-liquid-theory parameters, which are being experimentally measured these days.

V. YUKAWA INTERACTION MODEL AND DISCUSSION OF THE RESULTS

We will consider the effective interaction potential between the electrons to be of the Yukawa form, which enables us to perform all the calculations explicitly in terms of one parameter, the screening parameter.[1] Thus,

$$V_s(|\vec{x}-\vec{x}_1|)=1/[x+x_1-2(xx_1)^{1/2}\cos\theta_{xx_1}+\xi^2], \tag{30}$$

and so,

$$\frac{V_s^{(l)}(x;\,x_1)}{2l+1}=\frac{1}{2(xx_1)^{1/2}}\,Q_l\left(\frac{x+x_1+\xi^2}{2(xx_1)^{1/2}}\right), \tag{31}$$

where Q_l is the usual associated Legendre function. We then obtain

$$\Gamma=1\Big/\left\{1-\frac{\alpha r_s}{\pi}\left[1-\frac{\xi^2}{4}\ln\left(1+\frac{4}{\xi^2}\right)\right]\right\};$$

$$Z_1=1\Big/\left\{1+\frac{\alpha r_s}{\pi}\left[\frac{1}{2}\left(1+\frac{\xi^2}{2}\right)\ln\left(1+\frac{4}{\xi^2}\right)-1\right]\right\}, \tag{32}$$

$$\frac{J^{(2)}}{I^{(0)}}=\frac{1}{72}\left(\frac{\alpha r_s}{\pi}\right)\left[6-\frac{36}{\xi^2+4}-\frac{32}{(\xi^2+4)^2}-\tfrac{3}{2}\xi^2\ln\left(1+\frac{4}{\xi^2}\right)\right], \tag{33}$$

and

$$\left[\frac{d}{dx}\left(V_s^{(0)}(1;\,x)-x^{-1/2}\,\frac{V_s^{(1)}(1;\,x)}{3}\right)\right]_{x=1}$$
$$=\frac{1}{2}\left[-1-\frac{\xi^2}{2(4+\xi^2)}+\left(\frac{1}{4}+\frac{3\xi^2}{8}\right)\ln\left(1+\frac{4}{\xi^2}\right)\right]. \tag{34}$$

Several features of this model may be of value and so we record them here:

(i) In the large ξ limit (i.e., short-range model), the vertex equation can be solved exactly and one has to observe that the coupling strength is now $\alpha r_s/\pi\xi^2$ which is held constant. In this limit, we have $Z_1=1$, but $[d(V_s^{(0)}-\frac{1}{3}x^{-1/2}V_s^{(1)})/dx]_{x=1}\simeq O(1/\xi^4)$, and so the variational result coincides *completely with the exact answer*, as is to be expected.

(ii) In the limit of Coulomb interactions, i.e., $\xi\to 0$, we observe that Z_1 goes to zero like

$$1\Big/\left[\left(\frac{\alpha r_s}{2\pi}\right)\ln(4/\xi^2)\right];$$

$$\left(\frac{2\alpha r_s}{\pi}\right)^2\left[\frac{d}{dx}\left(V_s^{(0)}-x^{-1/2}V_s^{(1)}/3\right)\right]^2_{x=1}$$

diverges like $(\alpha r_s/2\pi)^2[\ln(4/\xi^2)]^2$ so that the correction to the variational answer diverges like $(\alpha r_s/2\pi)\ln(4/\xi^2)$. *In the strictly Coulomb limit, one has therefore a divergence* for a fixed r_s (or k_F). A similar result is also obtained by Kleinman.[11] But Kleinman takes ξ^2 to be the Thomas-Fermi expression, i.e., $\xi^2=4\alpha r_s/\pi$ and claims that now there is no divergence, as $r_s\to 0$. Hence, as long as ξ is treated as an independent parameter not depending on the electron density, one has a divergence, while if it depends on r_s, this divergence disappears. Now $\xi\to 0$ implies in the Thomas-Fermi model for screening that $r_s\to 0$; and so, the only inference possible is that in the high-density limit, the corrections to the variational answer are again zero. Also, we may point out that Sham's result is not expected to be correct for low densities, where $r_s\to\infty$ (or $k_F\to 0$).

(iii) In Ref. 13, we have developed a self-consistent scheme to determine ξ^2. The argument runs as follows. We know that in the limit of zero q, $q^2\epsilon_L(q)$ gives ξ^2. We have computed $\epsilon_L(q)$ using a Yukawa interaction, and this new expression for it should be consistent with the above general result. Thus, we obtain[13]

$$\xi^2_{sc}=(4\pi/k_F^2)\lim_{q\to 0}\chi(q)=(4\pi/k_F^2)\,x^{(0)},$$

or

$$\xi^2_{sc}=\frac{4\alpha r_s}{\pi}\,\Gamma=\left(\frac{4\alpha r_s}{\pi}\right)\Big/\left\{1-\left(\frac{\alpha r_s}{\pi}\right)\times\left[1-\tfrac{1}{4}\xi^2_{sc}\ln\left(1+\frac{4}{\xi^2_{sc}}\right)\right]\right\}. \tag{35}$$

This may be rewritten in the form

$$\frac{\alpha r_s}{\pi}=(\tfrac{1}{4}\xi^2_{sc}/[1+\tfrac{1}{4}\xi^2_{sc}-\tfrac{1}{16}\xi^4_{sc}\ln(1+4/\xi^2_{sc})]. \tag{36}$$

This equation can be solved graphically to determine ξ^2_{sc} as a function of $\alpha r_s/\pi$. We observe that $\xi^2_{sc}\to 0$ only if $\alpha r_s/\pi\to 0$, i.e, only in the very high-density limit. Thus the Thomas-Fermi relationship is reliable only for very high density of the electron gas. Also, for $\xi^2\to\infty$, we observe the right-hand side of Eq. (36) becomes proportional to $\frac{2}{3}(\xi^2_{sc}/4)$. Thus in the low-density limit, the screening, though similar to the Thomas-Fermi expression, is different by a factor $\frac{3}{2}$. This is not a reliable answer since one expects for very low densities zero screening.

It is worth noting that $\Omega(k_F)\equiv-(k_F^3/\pi^2)g_{xc}^{(2)}$, calculated using our formalism, vanishes in the two extreme limits of $\pi k_F\to 0$ (low density) and $\pi k_F\to\infty$ (high density) in contrast with the result of Kleinman.[11] Kleinman obtained a constant value for $\Omega(k_F)$ for $\pi k_F\to\infty$ (high density). To check if this would obtain in our formalism too, if we took ξ^2 to be the Thomas-Fermi value $4\alpha r_s/\pi$, we made a similar asymptotic calculation from our expression for $\Omega(k_F)$, and we found this to vanish. For $\pi k_F\to\infty$, we have the extreme high-density limit where Sham's procedure[10] is certainly valid, as is

also evident from our analysis. Sham found that $\Omega(k_F) \to 0$ for $\pi k_F \to \infty$, and we are in agreement with him. For $\pi k_F \to 0$ (low density), we agree with Kleinman's result that $\Omega \to 0$ again, whereas Sham's result is divergent but inappropriate in this limit. In view of the various comments made on the Yukawa model, it sppears a calculation of $\Omega(k_F)$ with a more realistic momentum-dependent interaction potential should be made. In Fig. 1 we have displayed our results. It is found that the variational results are in excellent agreement with the exact results for both high and low densities, except the position and height of the maximum are slightly different. We have therefore displayed only the exact results for the self-consistent and Thomas-Fermi schemes for ξ^2. The other contrasting features with the results of Sham and Kleinman are evident from this figure. A numerical table of Ω vs πk_F may be obtained on request, from either of the authors.

Professor Kleinman has informed one of us (A.K.R.) that he has now verified that his Ω also vanishes for $\pi k_F \to \infty$, in agreement with Sham and our result. The general shape of his curve of Ω vs πk_F now is the same as ours but seems to have some *small* differences in detail. We thank Professor Kleinman for informing us of his new finding and for pointing out a sign error in the definition of Ω in terms of $g_{xc}^{(2)}$. We must mention that we have also come across a paper by Geldart *et al.* [Solid State Commun. 16, 243 (1975)], where a rigorous analysis of the $g_{xc}^{(2)}$ is also made, and their conclusions in certain limits seem to be in general agreement with ours. These authors do not give a graph of Ω vs πk_F.

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