Bounds for Some Equilibrium Properties of an Electron Gas*

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Upper and lower bounds are derived for the average potential energy and Helmholtz free energy of an electron gas with uniform positive background. In the ground-state limit, upper and lower bounds are given for the average kinetic energy, average potential energy, and total ground-state energy. Inequalities are derived for the static form factor $S(k)$ and wave-number–dependent dielectric function $\epsilon(k, 0)$, making use of exact sum rules for the Fourier-transformed density-density commutator and of the assumption that $S(k) \leq 1$. Comparison is made with the exact behavior of these quantities for small k. The sum rules are used to construct an approximate nonlinear integral equation for the ground-state static form factor of the electron gas.

I. INTRODUCTION

In this paper we derive exact bounds for some equilibrium properties of an electron gas with uniform positive background. Mermin[1] recently derived exact lower bounds for this system in the classical limit, showing that the internal energy, Helmholtz free energy, and Fourier-transformed static form factor $S(\vec{k})$ are all bounded below by their Debye-Hückel values. In our discussion of the wave-number–dependent quantities $S(\vec{k})$, and $\epsilon(\vec{k}, 0)$ (the static dielectric function) particular emphasis is placed on the comparison of the bounds obtained with the exact behavior of these quantities for small k.

Starting with a variational property of the free energy (Sec. II), we first derive upper and lower bounds (valid for all temperatures and densities) for the Helmholtz free energy and potential energy, respectively. At temperature $T = 0$, we give both upper and lower bounds in terms of the dimensionless parameter r_s (defined by $\frac{4}{3}\pi r_s^3 a_0^3 = 1/\rho$, where a_0 is the Bohr radius and ρ the number density) for each of the quantities: average kinetic energy, average potential energy, and total ground-state energy. Most of these latter bounds are not new. However, since they apply to the entire range of r_s values and since the calculation of the above quantities in the high,[2] intermediate,[3] and low density[4] regions is still one of considerable interest, the results of Sec. II may be looked upon as providing a summary of simple criteria against which any approximation might be tested.

In Sec. III we review the relation of the quantities $S(\vec{k})$ and $\epsilon(\vec{k}, 0)$ to the basic spectral function $\chi''(\vec{k}, \omega)$ defined as the Fourier transform of the time-dependent density-density commutator. This function satisfies several exact sum rules[5,6] which form the basis for the inequalities obeyed by $S(\vec{k})$ and $\epsilon(\vec{k}, 0)$. The latter are cited in Sec. IV and compared with the known exact behavior of $S(\vec{k})$ and $\epsilon(\vec{k}, 0)$ for small k. The derivation of these inequalities (Sec. V) makes use of two general inequalities for the moment sums of $\chi''(\vec{k}, \omega)$ which are due to Mihara and Puff[7] and Bogoliubov,[8,9] respectively, and is based on the assumption that $S(\vec{k}) \leq 1$ in the homogeneous electron gas.

Finally, in Sec. VI we derive an approximate nonlinear integral equation for the static form factor of the ground-state electron gas. This integral equation is analogous to the one recently derived by Mihara and Puff[7] for ground-state He^4 using various moments of $\chi''(\vec{k}, \omega)$. The equation predicts the exact form of $S(\vec{k})$ for small k and leads to a convergent result for the radial distribution function evaluated at the origin. It also reproduces the free-fermion form factor $S_0(\vec{k})$ when the interaction is switched off.

II. BOUNDS FOR FREE ENERGY AND GROUND-STATE ENERGY

We consider a system of N electrons in a cubical box of volume Ω (with periodic boundary conditions), filled uniformly with positive background charge of density $|e|N/\Omega = |e|\rho$. The Hamiltonian H of the system will be written

$$H = H_0 + V, \tag{2.1}$$

where H_0 is the kinetic energy operator of the electrons, and

$$V = \frac{1}{2\Omega} \sum_{\vec{k}\neq 0} v(k)[\rho(\vec{k})\rho(-\vec{k}) - N] \tag{2.2}$$

is the potential energy operator of the system electrons plus background.[2] $\rho(\vec{k})$ and $v(k)$ are, respectively, the Fourier transforms of the number density operator $\rho(\vec{r}) = \sum_i \delta(\vec{r} - \vec{r}_i)$ and of the Coulomb potential $v(r) = e^2/r$; they are given by

$$\rho(\vec{k}) = \sum_i e^{-i\vec{k}\cdot\vec{r}_i}\,, \tag{2.3}$$

$$v(k) = 4\pi e^2/k^2\,. \tag{2.4}$$

The $\vec{k}=0$ Fourier component does not enter into the sum over $\vec{k}$ in (2.2) because this part of the interaction is cancelled by the contribution to the potential energy coming from the field of the uniform positive charge.

In a canonical ensemble of systems (identical to the one above) at temperature T, the Helmholtz free energy F and entropy S are given by

$$F = -k_B T \ln \mathrm{Tr} e^{-\beta H} = \langle H\rangle - TS, \tag{2.5}$$

where $\beta = 1/(k_B T)$, and the average value of an operator A is defined by

$$\langle A\rangle = \mathrm{Tr}\, e^{-\beta H} A/\mathrm{Tr}\, e^{-\beta H}\,. \tag{2.6}$$

It is generally accepted (even though a rigorous proof does not exist) that the minimum potential energy is realized when the particles are arranged in a static lattice configuration.[10] Denoting the potential energy of the static lattice by V_{st} we have

$$\langle V\rangle \geq V_{st}\,. \tag{2.7}$$

Of the lattice types thus far considered for a system of point charges in a uniform background of opposite charge, the bcc lattice has been found to have the lowest energy; for this lattice, V_{st} has the value[11] $V_{st}/N = -1.820\, e^2/a$, where $a = (2/\rho)^{1/3}$ is the side of the cube.

Together with (2.7) we shall obtain the inequalities

$$V_{st} \leq \langle V\rangle \leq \langle V\rangle_0\,, \tag{2.8}$$

$$F_0 + V_{st} \leq F \leq F_0 + \langle V\rangle_0\,, \tag{2.9}$$

$$F_0 \equiv \langle H_0\rangle_0 - TS_0 \leq \langle H_0\rangle - TS \leq F_0 + \langle V\rangle_0 - V_{st}\,. \tag{2.10}$$

The unperturbed quantities F_0, S_0, and $\langle A\rangle_0$ are defined by replacing H by the free-particle Hamiltonian H_0 in the definitions (2.5) and (2.6). The inequality $F \leq F_0 + \langle V\rangle_0$ means that the free energy of the system is bounded above by its value for a system of noninteracting electrons plus the first-order perturbation-theoretic value for the average interaction energy. This upper bound for F has already been obtained by many authors.[12] We emphasize the fact that the above inequalities are valid generally,[13] whether the equilibrium state of the system is uniform or crystalline.

The proof of the above relations makes use of the facts that F and F_0 can be written in terms of their respective density matrices P and P_0 as

$$F = \mathrm{Tr} P(H + k_B T \ln P)\,, \tag{2.11a}$$

$$F_0 = \mathrm{Tr} P_0(H_0 + k_B T \ln P_0)\,, \tag{2.11b}$$

where $$P = e^{-\beta H}/\mathrm{Tr} e^{-\beta H}\,, \tag{2.12}$$

and P_0 is similarly defined by replacing H by H_0 on the right-hand side of (2.12). The above forms for the density matrices guarantee that the free energies F and F_0 for the systems with Hamiltonians H and H_0, respectively, are minimized with respect to all Hermitian matrices with unit trace. Thus, if trial density matrices P_1 and P_2 are substituted in place of P and P_0 on the right-hand side of Eqs. (2.11a) and (2.11b) we have[14]

$$F \leq \mathrm{Tr} P_1(H + k_B T \ln P_1)\,, \tag{2.13}$$

$$F_0 \leq \mathrm{Tr} P_2(H_0 + k_B T \ln P_2)\,. \tag{2.14}$$

The choice $P_1 = P_0$ in (2.13) then gives the upper bound in (2.9), while the choice $P_2 = P$ in (2.14) yields $F_0 \leq \langle H_0\rangle - TS$. The upper bound for $\langle V\rangle$ in (2.8) results from these last two bounds and the definitions (2.1) and (2.5). Finally, the bounds $F_0 + V_{st} \leq F$ and $\langle H_0\rangle - TS \leq F_0 + \langle V\rangle_0 - V_{st}$ result from (2.7) and the other bounds derived above.

An important quantity in the study of the electron gas is the static form (or structure) factor $S(\vec{k})$ defined by

$$S(\vec{k}) = \frac{1}{N}\langle \rho(\vec{k})\rho(-\vec{k})\rangle\,, \tag{2.15a}$$

or, alternatively,

$$S(\vec{k}) = \frac{1}{N}\int d\vec{r}\, d\vec{r}\,' e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')}\langle \rho(\vec{r})\rho(\vec{r}\,')\rangle\,. \tag{2.15b}$$

At $\vec{k}=0$, $S(\vec{k})$ has the value N. From $S(\vec{k})$ the radial distribution function $g(\vec{r})$ is determined via

$$\rho[g(\vec{r}) - 1] = \frac{1}{\Omega}\sum_{\vec{k}\neq 0}[S(\vec{k}) - 1]\, e^{i\vec{k}\cdot\vec{r}}\,. \tag{2.16}$$

Furthermore, the average potential energy per particle is given in terms of $S(\vec{k})$ by[15]

$$\frac{\langle V\rangle}{N} = \frac{1}{2\Omega}\sum_{\vec{k}\neq 0} v(k)[S(\vec{k}) - 1]\,. \tag{2.17}$$

The quantity $\langle V\rangle_0$ reads

$$\frac{\langle V\rangle_0}{N} = \frac{1}{2\Omega}\sum_{\vec{k}\neq 0} v(k)[S_0(\vec{k}) - 1]\,, \tag{2.18}$$

where $S_0(\vec{k})$ is the static form factor of the free Fermi gas.[16] At $T=0$, $S_0(k)$ has the well-known value[17]

$$\begin{aligned} S_0(\vec{k}) &= N, && k = 0\\ S_0(\vec{k}) &= 3k/4k_F - k^3/16k_F^3\,, && 0 < k < 2k_F,\\ S_0(\vec{k}) &= 1\,, && 2k_F < k, \end{aligned} \tag{2.19}$$

where $k_F=(3\pi^2\rho)^{1/3}$ is the Fermi wave number. Substituting (2.4) and (2.19) into (2.18) we obtain

$$\langle V\rangle_0/N=-3e^2k_F/4\pi\ . \tag{2.20}$$

The right-hand side of (2.20) is the exchange energy evaluated in the Hartree-Fock approximation; it represents the expectation value of V over a Slater determinant of single-particle plane-wave states filling the Fermi sphere of radius k_F.[17]

With energies expressed in rydbergs (1 Ry $=e^2/(2a_0)=13.60$ eV) the relations (2.8)–(2.10) at $T=0$ imply the bounds for $\langle V\rangle/N$, E/N (the ground-state energy per particle), and $\langle H_0\rangle/N$ [the average kinetic energy per particle (hereafter denoted by $\langle \mathrm{KE}\rangle$)][18]:

$$-\frac{1.792}{r_s}\leq\frac{\langle V\rangle}{N}\leq-\frac{0.916}{r_s}\ , \tag{2.21}$$

$$\frac{2.210}{r_s^2}-\frac{1.792}{r_s}\leq\frac{E}{N}\leq\frac{2.210}{r_s^2}-\frac{0.916}{r_s}\ , \tag{2.22}$$

$$\frac{2.210}{r_s^2}\leq\langle \mathrm{KE}\rangle\leq\frac{2.210}{r_s^2}+\frac{0.876}{r_s}\ . \tag{2.23}$$

(2.22) can be written in an equivalent form[19] in terms of the correlation energy $E_c=E-E_{\mathrm{HF}}$:

$$-0.876/r_s\leq E_c/N\leq 0. \tag{2.22'}$$

As is well known, a lowering of the Hartree-Fock energy results for $r_s>5.46$, by having all spins in ferromagnetic alignment.[17] Because of the exclusion principle, the electrons in this case occupy all states up to a wave number $k_\dagger=(6\pi^2\rho)^{1/3}=2^{1/3}k_F$, so that a new upper bound results:

$$\frac{1}{N}E_{HF\dagger}=\frac{3}{5}\frac{\hbar^2k_\dagger^2}{2m}-\frac{3e^2k_\dagger}{4\pi}=\Delta_{\mathrm{ferr}}(r_s)+\frac{1}{N}E_{HF}\ , \tag{2.24}$$

where[17]

$$\Delta_{\mathrm{ferr}}(r_s)=\frac{2.210}{r_s^2}(2^{2/3}-1)-\frac{0.916}{r_s}(2^{1/3}-1)$$

$$=\frac{1.296}{r_s^2}-\frac{0.238}{r_s}\ \mathrm{Ry} \tag{2.25}$$

is negative for $r_s>5.46$. Hence, for such r_s we obtain improved upper bounds for $\langle V\rangle/N$, E/N, and $\langle \mathrm{KE}\rangle$ by adding $\Delta_{\mathrm{ferr}}(r_s)$ to the right-hand side of (2.21)–(2.23), respectively.

We emphasize the fact that the above bounds apply to the entire region of r_s values, whether the equilibrium state is uniform or crystalline; paramagnetic, ferromagnetic, or antiferromagnetic.[20]

In the classical limit the quantity $\langle V\rangle_0$ vanishes [since in this case $S_0(k)=1$] and (2.9) implies that F is bounded above by its value F_0 for an ideal gas at the same temperature and density. Together with the Debye-Hückel lower bounds[1] we have

$$-N^{\frac{1}{2}}e^2k_D\leq\langle V\rangle\leq 0\ , \tag{2.26}$$

$$F_0-N^{\frac{1}{3}}e^2k_D\leq F\leq F_0\ , \tag{2.27}$$

where $k_D^2=4\pi\rho e^2/(k_BT)$. It is evident that for large T the Debye-Hückel lower bounds provide an improvement over the lower bounds in (2.8) and (2.9).

III. TIME-DEPENDENT CORRELATIONS AND SUM RULES

In contrast to the relations derived thus far, where no assumption about the nature of the equilibrium state has been made, the relations to be derived in what follows will be valid only for the homogeneous electron gas, i.e., we henceforth assume our system to have translational and rotational invariance. We then have $\langle\rho(\vec{r})\rangle=\rho$ and the functions $S(\vec{k})$ and $g(\vec{r})$ will depend only on the magnitudes k and r, respectively.

The basic correlation function we consider is the time-dependent density-density commutator and its Fourier transform, the spectral function $\chi''(\vec{k},\omega)$. This latter quantity is defined by[5]

$$\chi''(\vec{k},\omega)=\int d\vec{r}\int_{-\infty}^{\infty}dt\,e^{-i\vec{k}\cdot\vec{r}+i\omega t}\tfrac{1}{2}\langle[\rho(\vec{r},t),\rho(0,0)]\rangle$$

$$=\int_{-\infty}^{\infty}dt\,e^{i\omega t}(1/2\Omega)\langle[\rho(\vec{k},t),\rho(-\vec{k},0)]\rangle, \tag{3.1}$$

where $\rho(\vec{k},t)=e^{itH}\rho(\vec{k},0)e^{-itH}$ is the Fourier transform of the number density operator $\rho(\vec{r},t)$ in the Heisenberg picture and where the average is defined as in (2.6). The product $\chi''(\vec{k},\omega)\times(1-e^{(-\beta\hbar\omega)})^{-1}$ is the density fluctuation spectrum measurable in inelastic electron scattering experiments; $\hbar\vec{k}$ is the momentum transfer and $\hbar\omega$ the energy loss of the scattered electron.

As a consequence of its definition, together with the spatial invariance of the system, the quantity $\chi''(\vec{k},\omega)$ is real and has the properties[5]

$$\chi''(\vec{k},\omega)=-\chi''(\vec{k},-\omega)=\chi''(k,\omega),\quad \omega\chi''(k,\omega)\geq 0\ . \tag{3.2}$$

The static form factor is related to $\chi''(k,\omega)$ by the equation[6]

$$\rho S(k)=\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\chi''(k,\omega)\coth(\tfrac{1}{2}\beta\hbar\omega)\ , \tag{3.3}$$

which may be looked upon as a sum rule for $\chi''(k,\omega)$.

An important quantity obtainable from $\chi''(k,\omega)$ is the density-density response function; the latter measures the linear response of the system to an external potential acting on the density. The Fourier transform of this quantity, $\chi(k,\omega)$,

is given by[5]

$$\chi(k,\omega)=\chi'(k,\omega)+i\chi''(k,\omega)\,, \tag{3.4}$$

which represents the boundary value as z approaches ω on the real axis from above, of the analytic function of z

$$\chi(k,z)=\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\,\frac{\chi''(k,\omega')}{\omega'-z}\,. \tag{3.5}$$

The static wave-number–dependent susceptibility $\chi(k)$ is defined as the inverse first moment of $\chi''(k,\omega)$:

$$\chi(k)=\chi(k,0)=\chi'(k,0)=\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\,\frac{\chi''(k,\omega')}{\omega'}\,; \tag{3.6}$$

its physical significance is stated below.

Other exact relations satisfied by $\chi''(k,\omega)$ will be needed. The first is the well-known longitudinal f sum rule[5]

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi''(k,\omega)=\frac{\rho\hbar k^2}{m}\equiv 2\rho\omega_0(k)\,, \tag{3.7}$$

which holds whenever the interparticle forces are velocity independent, and the second is the ω^3-moment sum rule[6]

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega^3\chi''(k,\omega)=2\rho\omega_0(k)\omega_3^2(k)\,, \tag{3.8}$$

where $$\omega_3^2(k)=\omega_0^2(k)+4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar+Q(k)\,. \tag{3.9}$$

The function $Q(k)$ is defined as[21]

$$Q(k)=\frac{1}{Nm}\sum_{i,j=1}^{N}\langle e^{-i\vec{k}\cdot(\vec{r}_i-\vec{r}_j)}(\hat{\vec{k}}\cdot\nabla_i)(\hat{\vec{k}}\cdot\nabla_j)V\rangle\,, \tag{3.10}$$

where $\hat{\vec{k}}$ is a unit vector in the direction of $\vec{k}$ and $\nabla_i=\partial/\partial\vec{r}_i$. When the form of V given by (2.2) is used in (3.10), $Q(k)$ can be written

$$Q(k)=\frac{1}{m\Omega}\sum_{\vec{q}\neq 0}(\hat{\vec{k}}\cdot\vec{q})^2 v(q)[S(\vec{q}+\vec{k})-S(\vec{q})]\,. \tag{3.11}$$

In expressing the above sum as an integral over q space, we must first separate the contribution $Nk^2v(k)/(m\Omega)=4\pi\rho e^2/m=\omega_{p1}^2$ (ω_{p1} being the plasma frequency) coming from the term $\vec{q}=-\vec{k}$.

We then obtain $$Q(k)=\omega_{p1}^2+I(k)\,, \tag{3.12}$$

where, explicitly

$$I(k)=\frac{1}{m}\int\frac{d^3q}{(2\pi)^3}\{[\hat{\vec{k}}\cdot(\vec{q}+\vec{k})]^2\times v(\vec{q}+\vec{k})-(\hat{\vec{k}}\cdot\vec{q})^2 v(q)\}[S(q)-1]\,. \tag{3.13}$$

The response of the electron gas to a longitudinal external field may equivalently be described in terms of the longitudinal dielectric function[2] $\epsilon(k,\omega)$. The latter is related to $\chi(k,\omega)$ by

$$[\epsilon(k,\omega)]^{-1}=1-(4\pi e^2/\hbar k^2)\chi(k,\omega)\,, \tag{3.14}$$

from which we obtain the relation between $\chi(k)$ and the static dielectric function $\epsilon(k,0)$:

$$[\epsilon(k,0)-1]/\epsilon(k,0)=(4\pi e^2/\hbar k^2)\chi(k)\,. \tag{3.15}$$

Physically, the ratio on the left-hand side [or, equivalently, $\chi(k)$] is a measure of the screening in the electron gas of a static charge disturbance of wave number k.[2,22]

IV. INEQUALITIES SATISFIED BY $S(k)$, $\chi(k)$, AND $\epsilon(k,0)$

As a consequence of the relations (3.2) and (3.6) it is noted that $\chi(k)$ is a positive quantity, implying[23]

$$[\epsilon(k,0)-1]/\epsilon(k,0)\geq 0\,. \tag{4.1}$$

From this it follows that either $\epsilon(k,0)\leq 0$, or $\epsilon(k,0)\geq 1$. It has been suggested[23] and recently verified by actual calculation[24] that the case $\epsilon(k,0)\leq 0$ is realized in the low-density electron lattice[25] (the so-called Wigner lattice). Since the latter represents a spatially noninvariant state of the system, it violates the assumption made in Sec. III and will not be dealt with subsequently. The other possibility, $\epsilon(k,0)\geq 1$, is therefore taken to apply to the homogeneous electron gas.[26]

A stronger bound for $\epsilon(k,0)$ is contained in the following inequalities to be derived in Sec. V on the basis of the definitions and sum rules of Sec. III and the additional assumption $S(k)\leq 1$:

$$S(k)\geq\left(\frac{\omega_0^2(k)}{\omega_{p1}^2+4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar+\omega_0^2(k)}\right)^{1/2}, \tag{4.2}$$

$$\chi(k)\geq\frac{2\rho\omega_0(k)}{\omega_{p1}^2+4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar+\omega_0^2(k)}\,, \tag{4.3}$$

$$\beta\hbar\rho S(k)\geq\chi(k)\,, \tag{4.4}$$

$$\epsilon(k,0)\geq 1+\frac{\omega_{p1}^2}{\omega_0(k)[4\langle \mathrm{KE}\rangle/\hbar+\omega_0(k)]}\,. \tag{4.5a}$$

The last relation follows from (4.3) and (3.15).

At $T=0$ we also have the inequality

$$\omega_0(k)\chi(k)\geq 2\rho[S(k)]^2\,, \tag{4.5b}$$

or, stated in terms of $\epsilon(k,0)$ using (3.15),

$$[\epsilon(k,0)-1]/\epsilon(k,0)\geq[\omega_{p1}S(k)/\omega_0(k)]^2\,, \tag{4.5c}$$

$$\epsilon(k,0)\geq\{1-[\omega_{p1}S(k)/\omega_0(k)]^2\}^{-1}\,. \tag{4.5d}$$

The last relation implies that for large k

$$\epsilon(k,0)\geq\{1-[\omega_{p1}/\omega_0(k)]^2\}^{-1}\,. \tag{4.5e}$$

A characteristic feature of the interacting elec-

tron gas is that for long wave lengths its properties are determined entirely by the plasma oscillations.[2] As a consequence, in the limit of small k the quantities $S(k)$, $\chi(k)$, and $\epsilon(k, 0)$ are given by the exact relations valid at any temperature:

$$S(k) = (\hbar k^2/2m\omega_{pl})\coth\tfrac{1}{2}\beta\hbar\omega_{pl}\,, \quad k\to 0 \tag{4.6}$$

$$\chi(k) = \hbar k^2/4\pi e^2\,, \quad k\to 0 \tag{4.7}$$

$$\epsilon(k, 0) = 1 + \frac{\omega_{pl}^2}{s^2k^2}\,, \quad k\to 0\,. \tag{4.8}$$

Here s is the isothermal sound velocity related to the isothermal compressibility K_T and Fermi level μ by

$$\frac{1}{ms^2} = \rho K_T = \frac{1}{N}\left(\frac{\partial N}{\partial \mu}\right)_{T,\Omega}\,. \tag{4.9}$$

The relation (4.8) is often expressed in terms of the inverse screening length k_s defined by $k_s = \omega_{pl}/s$. Equations (4.7) and (4.8) are expressions of the fact that in the long-wave-length limit the electron gas exhibits perfect screening.[2]

The limiting forms (4.6) and (4.7) follow from the definitions (3.3), (3.6), and the form of the spectral function $\chi''(k, \omega)$ for small k[2,9]:

$$\chi''(k, \omega) = \frac{\pi\rho\hbar k^2}{2m\omega(k)}\{\delta[\omega-\omega(k)] - \delta[\omega+\omega(k)]\}\,,$$
$$k\to 0, \quad \text{with } \omega(k)\to\omega_{pl} \quad \text{for } k\to 0\,. \tag{4.10}$$

This $\chi''(k, \omega)$ exhausts both the sum rules (3.7) and (3.8) (for $k\to 0$).

Comparing (4.7) with (4.3) we see that for small k the inequality becomes an equality. In the ground-state limit ($\beta\to\infty$) the coth factor in (4.6) is 1 and $S(k)$ is given by

$$S(k) = \hbar k^2/2m\omega_{pl}\,, \quad k\to 0, \quad T=0\,; \tag{4.11}$$

comparison with (4.2) shows that in this limit that inequality also becomes an equality. On the other hand, (4.4) reduces to a trivial statement at $T=0$.

In the classical limit, $\coth(\frac{1}{2}\beta\hbar\omega_{pl}) = 2/(\beta\hbar\omega_{pl})$ and (4.6) implies that

$$S(k) = k^2/k_D^2\,, \quad k\to 0, \tag{4.12}$$

showing that for small k (4.4) becomes an equality. The result (4.12) shows that the inequality derived by Mermin,[1]

$$S(k) \geq k^2/(k_D^2 + k^2)\,, \tag{4.13}$$

also becomes an equality for small k.[27]

Comparison of (4.5) and (4.8) leads to an inequality for the isothermal sound velocity in the electron gas[28]:

$$\tfrac{1}{2}ms^2 \leq \langle \mathrm{KE}\rangle\,, \tag{4.14}$$

or, stated equivalently in terms of the exact inverse screening length k_s:

$$k_s^2/k_{FT}^2 \geq 5\langle \mathrm{KE}\rangle_0/9\langle \mathrm{KE}\rangle, \tag{4.15}$$

where $k_{FT}^2 = 6\pi\rho e^2/(\frac{5}{3}\langle \mathrm{KE}\rangle_0)$ coincides with the square of the Fermi-Thomas wave number at $T=0$.

V. DERIVATION OF INEQUALITIES FOR $S(k)$ AND $\chi(k)$

The derivation of the inequalities (4.2) and (4.3) makes use of two general inequalities due to Mihara and Puff[7] and Bogoliubov,[8,9] respectively. These have been derived making use of the relations (3.2), (3.3), and (3.6) and of the sum rules (3.7) and (3.8).

The inequality of Mihara and Puff reads

$$\left(\frac{S(k)}{\omega_0(k)}\right)^2 \geq \int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi''(k, \omega)\Big/\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\omega^3\chi''(k, \omega)$$
$$= \frac{1}{\omega_3^2(k)}\,, \tag{5.1}$$

and that of Bogoliubov, applied to the density-density correlations, reads

$$\beta\hbar\rho S(k) \geq \chi(k) \geq \left(\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi''k, \omega\right)^2$$
$$\times\left(\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega^3\chi''(k, \omega)\right)^{-1} = \frac{2\rho\omega_0(k)}{\omega_3^2(k)}\,, \tag{5.2}$$

where $\omega_0(k)$ and $\omega_3(k)$ are defined by (3.7) and (3.9). Using the expressions (3.9) and (3.12), the above inequalities become

$$S(k) \geq \left(\frac{\omega_0^2(k)}{\omega_{pl}^2 + 4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar + \omega_0^2(k) + I(k)}\right)^{1/2}, \tag{5.3}$$

$$\chi(k) \geq \frac{2\rho\omega_0(k)}{\omega_{pl}^2 + 4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar + \omega_0^2(k) + I(k)}\,, \tag{5.4}$$

where $I(k)$ is defined by Eq. (3.13). If $I(k)$ can be shown to have the property $I(k)\leq 0$ for all k, the above inequalities will be strengthened by replacing $I(k)$ by its maximum value, zero, thus yielding (4.2)–(4.4). We now show that the inequality $I(k)\leq 0$ follows from a plausible assumption about the behavior of $S(k)$.

When (2.4) is substituted into (3.13) and the integration over solid angle is carried out, we obtain

$$I(k) = \frac{e^2}{m\pi}\int_0^{\infty} q^2\,dq[S(q) - 1]J(q, k)\,, \tag{5.5}$$

where $J(q, k) = \frac{5}{6} - \frac{q^2}{2k^2} + \frac{k}{8q}\left(\frac{q^2}{k^2} - 1\right)^2 \ln\left(\frac{q+k}{q-k}\right)^2$ (5.6)

is a function only of the ratio q/k, positive for all values of its arguments and monotonically de-

creasing. Further properties of the functions $J(q, k)$ and $I(k)$ are given in the Appendix.

For small k we have seen that $S(k)$ goes as k^2; consequently, the integrand in (5.5) is negative for small q values. Now, a sufficient condition (but by no means a necessary one) for $I(k) \leq 0$ is that the integrand in (5.5) be negative, i.e., $S(k) \leq 1$ for all k. To see whether this condition obtains, we note that since $S(k)$ must tend to 1 for large k, it follows that for intermediate values of k the function $S(k)$ can either increase monotonically from 0 to 1 as k increases, or else it can have a peak for some $k = k_m$. An indication of the behavior of $S(k)$ in a normal electron gas is revealed by its value in the RPA[29] at $T = 0$; the latter (see Fig. 1) shows that no such peak exists and that $S_{\mathrm{RPA}}(k)$ is monotonically increasing. We assume this property also holds for the exact $S(k)$ in a homogeneous electron gas with uniform background. With the assumption of monotonicity[30] for $S(k)$, the integrand in (5.5) is negative for all q and the property $I(k) \leq 0$ follows. As shown in the Appendix, this property holds in the limit of small and large k even without the assumption of monotonicity.

The plausibility of the above assumption is strengthened by a further observation: First, it follows from Eqs. (4.6) and (2.19) that for small k, $S(k) \leq S_0(k)$. Figure 1 shows that $S_{\mathrm{RPA}}(k) \leq S_0(k)$ for all k, tempting the conjecture that the actual $S(k) \leq S_0(k)$ for all k. This conjecture is supported by the inequality $\langle V \rangle \leq \langle V \rangle_0$ proved in (2.8), which follows trivially if $S(k) \leq S_0(k)$. Together with (4.2) we would then have both an upper and lower bound for $S(k)$:

$$\frac{\omega_0(k)}{[\omega_{p1}^2 + 4\omega_0(k)\langle \mathrm{KE} \rangle/\hbar + \omega_0^2(k)]^{1/2}} \leq S(k) \leq S_0(k), \quad (5.7)$$

where $S_0(k)$ is defined by Eq. (2.19) at $T = 0$.

Finally, we prove the inequalities (4.5b)–(4.5e) at $T = 0$. From (3.3) $S(k)$ is in this case given by

$$\rho S(k) = \int_0^\infty \frac{d\omega}{\pi} \chi''(k, \omega). \quad (5.8)$$

Using the fact that $\omega\chi''(k, \omega) \geq 0$, we can write the inequality

$$\int_0^\infty \frac{d\omega}{\pi} \frac{\chi''(k, \omega)}{\omega} [\omega - \omega(k)]^2 \geq 0, \quad (5.9)$$

valid for any real function $\omega(k)$. Expanding the above integrand and carrying out the integrations term by term, using Eqs. (5.8), (3.6), and (3.7) we obtain

$$\rho\omega_0(k) - 2\rho\omega(k)S(k) + \tfrac{1}{2}[\omega(k)]^2\chi(k) \geq 0, \quad (5.10)$$

for any $\omega(k)$. The choice $\omega(k) = \omega_0(k)/S(k)$ then leads directly to (4.5b). We note that this choice for $\omega(k)$ is precisely the Feynman excitation frequency[31] for a density fluctuation of wave vector k. [Using for $S(k)$ the exact form (4.11) for $k \to 0$, the Feynman excitation frequency gives, of course, the frequency of a long-wave-length plasma oscillation, i.e., $\omega(k \to 0) = \omega_{p1}$.[32]]

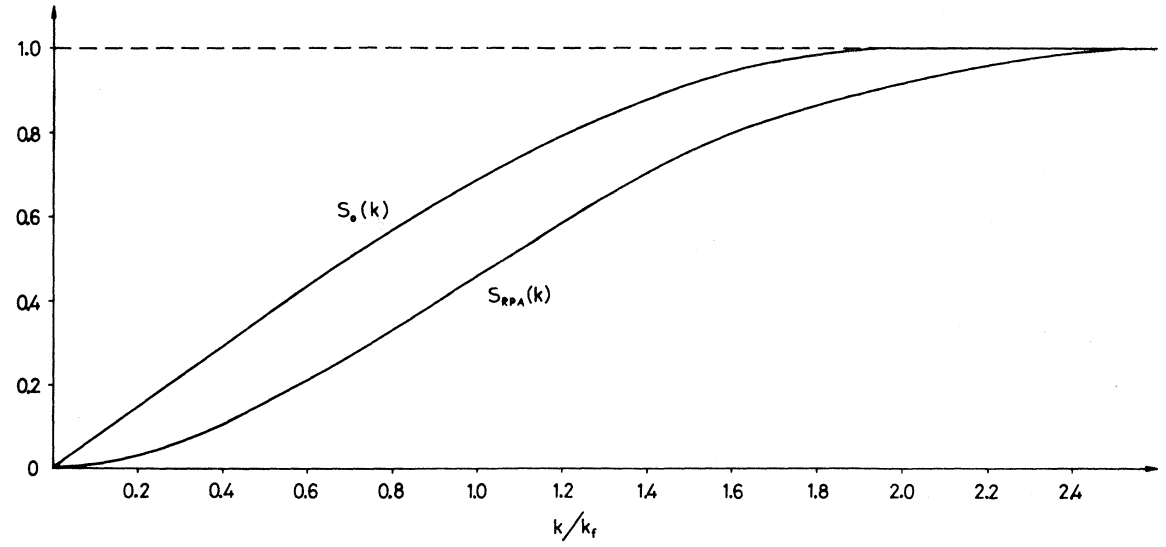

FIG. 1. Plot of the ground-state free-fermion static form factor $S_0(k)$ [Eq. (2.19)] and of the RPA value of $S(k)$ obtained by Glick and Ferrell (Ref. 29); the latter was calculated at a density corresponding to that of aluminum ($r_s \sim 2$).

VI. INTEGRAL EQUATION FOR $S(k)$

We now show how the inequalities derived in Sec. V can be used to construct a nonlinear integral equation for the static form factor of the ground-state electron gas. This equation is analogous to the one first derived by Mihara and Puff[7] (MP) for ground-state He^4; our derivation closely follows their work.

We have already seen that for small k, $S(k)$ is equal to its lower bound (lb) given by the right-hand side of (5.3), both behaving as $\hbar k^2/(2m\omega_{pl})$. Also, for large k, both $S(k)$ and its lb approach unity. [The fact that for $k\to 0$, $I(k)\to 0$ and for $k\to\infty$, $I(k)\to \text{const}\times\omega_{pl}^2$ are shown in the Appendix.] Hence, we might at first be tempted to define the integral equation for $S(k)$ by simply equating $S(k)$ to its lb (5.3). The resulting integral equation, written as $[\omega_0(k)/S(k)]^2=\omega_3^2(k)$ [with $\omega_3^2(k)$ given by (3.9), (3.12), and (5.5)] would then be equivalent to postulating the single-resonance ansatz (4.10) for all k, with $\omega(k)=\omega_0(k)/S(k)$, the Feynman excitation frequency. However, this integral equation leads to a difficulty at large k having its origin in the term $4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar$. Due to the latter, the right-hand side of (5.3) would predict that for large k, $S(k)$ approaches unity as $1-m\langle \mathrm{KE}\rangle/\hbar^2k^2)$, resulting in a $g(r)$ which is negatively divergent as $r\to 0$.

We follow MP in writing the inequality (5.3) as

$$[\omega_0(k)/S(k)]^2=\omega_3^2(k)-f(k)\,, \tag{6.1}$$

where $f(k)\geq 0$ for all k. To use (6.1) as an integral equation for $S(k)$, $f(k)$ must have as its leading term at large k the value $4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar$. The kinetic energy term is then eliminated from the equation at high k and a solution for which $g(r)$ is finite at the origin is possible. The simplest choice which satisfies the above requirements for $f(k)$ is then to take $f(k)=4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar$ for all k, leading to the integral equation

$$[\omega_0(k)/S(k)]^2=\omega_0^2(k)+\omega_{pl}^2+I(k)\,, \tag{6.2}$$

with $I(k)$ defined by (5.5). We note that this equation gives $S(k)=1$ when the coupling constant e^2 is put equal to zero. However, for any nonzero value of e^2, Eq. (6.2) gives the exact value of $S(k)$ for small k and leads to a finite value of $g(0)$.

The fact that for zero value of the coupling constant, Eq. (6.2) leads to the value $S(k)=1$ appropriate for noninteracting classical particles, rather than the value $S_0(k)$ [Eq. (2.19)] describing free fermions at zero temperature is not surprising, since it is well known that the single-resonance ansatz (4.10) is not valid for free fermions.

We now show how Eq. (6.2) may be improved so as to give a better account of the correlations implied by the fermion nature of our particles. We define the function $g(k)$ by writing

$$[\omega_0(k)/S(k)]^2=\omega_0^2(k)+\omega_{pl}^2+I(k)+g(k)\,, \tag{6.3}$$

where $g(k)\leq 4\omega_0(k)\langle \mathrm{KE}\rangle/\hbar$ for all k and in particular less than order k^2 for large k [this follows from the above restrictions on $f(k)$]. We now specify a form for $g(k)$ by simply demanding that Eq. (6.3) give the correct free-fermion form factor $S_0(k)$ when e^2 is set equal to zero. This leads to

$$g(k)=\omega_0^2(k)\left[\left(\frac{3}{4}\frac{k}{k_F}-\frac{1}{16}\frac{k^3}{k_F^3}\right)^{-2}-1\right],\quad 0\leq k\leq 2k_F$$

$$g(k)=0\,,\qquad 2k_F\leq k\,. \tag{6.4}$$

It is easily verified that this choice for $g(k)$ satisfies all the above requirements. For small k, $g(k)$ behaves as $\frac{80}{27}\omega_0(k)\langle \mathrm{KE}\rangle_0/\hbar$ (where $\langle \mathrm{KE}\rangle_0=3\hbar^2k_F^2/10\,\mathrm{m}$), then rises to a maximum and drops continuously to the value zero at $k=2k_F$. With the above choice for $g(k)$, Eq. (6.3) can finally be written in the form

$$[\omega_0(k)/S(k)]^2=[\omega_0(k)/S_0(k)]^2+\omega_{pl}^2+I(k)\,, \tag{6.5}$$

where $S_0(k)$ and $I(k)$ are defined by Eqs. (2.19) and (5.5), respectively. The above equation gives the exact value of $S(k)$ for small k, is consistent with the moment sum rules, leads to a finite value of $g(0)$, and takes into account the additional correlations arising from the exclusion principle.

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APPENDIX

Herein we give some further properties of the functions $J(q,k)$ and $I(k)$ defined by Eqs. (5.5) and (5.6). For $q/k<1$, $J(q,k)$ has the expansion

$$J(q,k)=\frac{4}{3}-\frac{4}{3}\frac{q^2}{k^2}+\frac{4}{15}\frac{q^4}{k^4}+\cdots\,, \tag{A1}$$

while for $q/k>1$, $J(q,k)$ can be written

$$J(q,k)=\frac{4}{15}\frac{k^2}{q^2}+\frac{4}{105}\frac{k^4}{q^4}+\cdots\,. \tag{A2}$$

At $q=k$, J has the value $\frac{1}{3}$.

Substituting (A1) into (5.5) we obtain the limiting value of $I(k)$ for $k\to\infty$:

$$I(k)=\frac{4e^2}{3m\pi}\int_0^\infty q^2\,dq[S(q)-1]=\tfrac{2}{3}\omega_{pl}^2[g(0)-1]\,, \tag{A3}$$

where $g(0)$ is the radial distribution function (2.16) evaluated at $r=0$. The part of $g(0)$ which is due to particles of parallel spin vanishes automatically on account of the exclusion principle. In the remaining part, contributed by particles of antiparallel spin, the dynamical correlation due to the Coulomb repulsion will act to prevent these particles from approaching too closely, with the result that $g(0)<1$, and hence that for large k, $I(k)<0$.

To obtain the behavior of $I(k)$ for small k, we first rewrite (5.5), changing the integration variable to $y=q/k$:

$$I(k)=\frac{e^2}{m\pi}\,k^3\int_0^\infty y^2\,dy[S(ky)-1]J(y)\ . \tag{A4}$$

To obtain the k dependence of the above integral for small k, we write the latter as $\int_0^\infty=\int_0^1+\int_1^\infty$. Making use of the known form of $S(yk)$ for small $k[S(yk)=\hbar k^2y^2/(2m\omega_{p1})]$, the part contributed by $\int_0^1$ is of the form $-a+bk^2$, where a and b are positive numbers; hence this part leads to an $I(k)\sim k^3$.

On the other hand, for $k\to 0$, the integral

$$\begin{aligned}\int_1^\infty y^2\,dy[S(ky)-1]J(y)&\to\tfrac{4}{15}\int_1^\infty dy[S(ky)-1]\\&=\tfrac{4}{15}\{\textstyle\int_0^\infty dy[S(ky)-1]-\int_0^1 dy[S(ky)-1]\}\\&\to(4/15k)\textstyle\int_0^\infty dq[S(q)-1]\ ,\end{aligned} \tag{A5}$$

so that to leading order in k(A4) becomes

$$I(k)=\frac{4}{15}\,\frac{k^2}{m}\,\frac{\langle V\rangle}{N}\ ,\quad k\to 0\ , \tag{A6}$$

where use has been made of the relation [(2.17)] between the average potential energy per particle and $S(k)$. Since $\langle V\rangle/N$ is negative, we have shown $I(k)$ negative also for small k, independent of the assumption of monotonicity.

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[12]It appears, for example, as Eq. (3) in R. B. Griffiths, J. Math. Phys. 5, 1215 (1964), where earlier references may be found. A more recent discussion of this inequality is found in H. S. Leff, Am. J. Phys. 37, 548 (1969).

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[14]The minimum property of the quantum-mechanical free energy was first proved by R. E. Peierls [Phys. Rev. 54, 918 (1938)].

[15]It is understood that in evaluating sums over k by integration over k space, the transition to the thermodynamic limit is always implied, i.e., $N,\Omega\to\infty$ with $N/\Omega=\rho$ and

$$(1/\Omega)\sum_{\mathbf{k}\neq 0}\to\int\frac{d^3k}{(2\pi)^3}\ .$$

[16]Reference 2, Chap. 2.

[17]D. Pines, *Elementary Excitations in Solids* (Benjamin, New York, 1963), Chap. 3.

[18]The bounds (2.21) and (2.22) are already implicit in Wigner's work on the correlation energy. See Ref. 17, pp. 89–95. The fact that $\langle$KE$\rangle$ is bounded below by its value for free electrons was shown by R. M. Mazo [J. Math. Phys. 8, 1546 (1967)].

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$$Q(k)=\frac{\rho}{m}\int d\vec{r}\,g(r)[1-\cos(\vec{k}\cdot\vec{r})](\hat{k}\cdot\nabla)^2v(r).$$

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[26]It has been observed (see Ref. 2, pp. 207, and Ref. 23, pp. 148) that $\epsilon(\vec{k},0)\leq 0$ implies the nonanalyticity of $\epsilon(\vec{k},\omega)$ in the upper half of the complex ω plane. By means of a physical argument, Pines and Nozières have

shown that if $\epsilon(\vec{k}, 0)$ is negative for some value of $\vec{k}$, the positive background will be unstable against the development of spontaneous density fluctuations of this wave vector. In the long-wave-length limit $\epsilon(\vec{k}, 0) \leq 0$ corresponds to a negative compressibility of the system electrons plus background. The fact that the low-density electron lattice state is one of negative compressibility has been pointed out by H. M. Van Horn [Phys. Rev. 157, 342 (1967)]. The conclusion reached by Pines and Nozières is that the condition $\epsilon(\vec{k}, 0) > 0$ [which implies the analyticity of $\epsilon(\vec{k}, \omega)$ in the upper half of the complex ω plane] follows from the requirement that the positive background be stable under the influence of the electron gas.

[27]For arbitrary k, the relations (4.3) and (4.4) in the classical limit imply $S(k) \geq k^2/(k_D^2 + 3k^2)$, which is a weaker statement than (4.13).

[28]This relation is verified in the noninteracting case at $T = 0$, where the sound velocity $s_0 = \hbar k_F/(\sqrt{3}m)$. Similarly, in the case of a low-density weakly interacting classical plasma, s^2 is given by the value $k_B T/m$, again in agreement with (4.14).

[29]A. J. Glick and R. A. Ferrell, Ann. Phys. (N.Y.) 11, 359 (1960).

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[31]R. P. Feynman, Phys. Rev. 94, 262 (1954). Although the Bijl-Feynman trial excited state $\rho(\vec{k})|\Phi_0\rangle$ (where $|\Phi_0\rangle$ is the exact ground state) was applied originally to He^4, this state works equally well as a trial excited state for a system of fermions. This follows from the fact that $\rho(\vec{k})|\Phi_0\rangle$ has the same symmetry property (symmetric or antisymmetric under exchange of particles) as $|\Phi_0\rangle$, and is orthogonal to it.

[32]The fact that the Feynman result is exact for Bose systems at $T = 0$ in the limit $k \to 0$ has been shown by A. Miller, D. Pines, and P. Nozières [Phys. Rev. 127, 1452 (1962)]. For the electron gas this follows from the form (4.10) of $\chi'(k, \omega)$.

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Anticorrelation in Two-Photon Attenuated Laser Beam

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The density matrix in the P representation of a beam of radiation amplified by a two-photon amplifier has been derived up to the lowest order in the time-dependent perturbation theory without placing any restriction on the population of the state of the atom. It is shown that a laser beam containing noise in addition to the harmonic signal exhibits anticorrelation after being passed through such an amplifier, if less than one-sixth of the total number of atoms are maintained in the excited state.

INTRODUCTION

Recently, there has been a good deal of discussion[1-6] on the correspondence between the newly developing quantum theory[6-9] of optical coherence and the older semiclassical theory.[3,4,8-12] The classical definition of coherence functions is identical with the quantum definition, if the weight functional $P(\{v_{\vec{k}}\})$ in the diagonal phase-space representation of the density operator is real, nonsingular, and non-negative. There also exist fields for which $P(\{v_{\vec{k}}\})$ takes negative values in some regions of the complex $v_{\vec{k}}$ planes. These fields do not have classical analogs. With such fields, lesser photon coincidences than the random background may be recorded in a Hanbury Brown–Twiss detector.[13] Radiation in a pure Fock state is an example of such fields. This effect, referred to as anticorrelation, has not so far been observed experimentally, because it is very unusual in practice to have well-defined numbers of photons and because the conventional sources of optical fields have non-negative values of $P(\{v_{\vec{k}}\})$ throughout the complex $v_{\vec{k}}$ planes.

Recently, the authors have shown[14] that the statistical nature of photons is changed after interaction with a one-photon oscillator. Photon oscillators can thus be used for producing optical fields with photon statistics different from those of conventional sources. In this paper, we shall show that it is possible to obtain an optical field which can exhibit anticorrelation from a laser beam, by passing it through a two-photon oscillator. This gives a practical method of observing anticorrelation with the help of ideal photodetection.[15]

DENSITY MATRIX OF OUTPUT RADIATION

Let us consider an atomic system interacting with a single-mode radiation field. The Hamiltonian of this system in Heisenberg representation can