LECTURES ON THE MANY-ELECTRON PROBLEM

By R. BROUT

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VOLUME X

PREFACE

This book originated in a course given by R. Brout at Cornell University on the many-body problem. The emphasis was on two subjects. The first was a detailed introduction to the linked cluster development of the many-body perturbation theory with parallel development in classical statistics, quantum adiabatic zero temperature theory, and finally quantum statistics. The second was a systematic development of the many-electron problem from the plasma point of view.

We decided to adhere in great measure to this plan. Thus the book is of an intensive rather than extensive character. It was felt of pedagogical necessity to go into great detail on the formal manipulations of perturbation theory. This formal theory is then given substance by detailed application to one problem of importance in physics.

It is with considerable regret that requirements of time and space have not permitted us to write further on some of the more elegant developments of many-body theory as well as on the beautiful applications that have been found in physical problems. We also mention here that the student will profit considerably by reading in parallel with the present work the excellent books of Pines and Thouless. These books are wider in scope than ours and consequently less detailed in development.

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CONTENTS

The Many-Body Problem in Classical Statistical Mechanics

1.1. Ursell-Mayer Cluster Expansion

The prototype of the classical many-body problem is the dilute non-ideal monatomic gas, whose macroscopic behavior is described by the Ursell-Mayer expansion. Aside from giving a simpler proof of this expansion than is usual, our purpose in presenting this theory is twofold. In the first place, the analysis of this problem exposes in a familiar context many features common to all many-particle systems. Secondly, the modification of this analysis required by the long range of the coulomb force sheds much light on the collective behavior of many-electron systems.

According to statistical mechanics all thermodynamic quantities may be calculated from the free energy F. In the classical canonical ensemble the free energy is given in terms of the *partition function* Z according to the relation

$$-\beta F = \log Z; \quad \beta \equiv 1/kT, \tag{1-1}$$

$$Z = \frac{1}{N!\, h^{3N}} \int dp\, dq \exp\left[-\beta H(p, q)\right]. \tag{1-2}$$

In these equations T is the absolute temperature, k is Boltzmann's constant; h is Planck's constant. The N-particle system is described by the Hamiltonian $H(p, q)$ where p stands for the $3N$ momenta $p_1, p_2, \ldots, p_N$ and q for the $3N$ coordinates $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N$. Thus, in Eq. (1-2): $dp = \prod_{j=1}^{N} d^3p_j$, $dq = \prod_{j=1}^{N} d^3r_j$. We suppose further that all particles have the same mass, m. Then for an ideal gas, $H = \Sigma\, \mathbf{p}_i^2/2m$, Eq. (1-2) becomes ($Z_{\text{ideal}} \equiv Z_0$)

$$Z_0 = \frac{\Omega^N}{N!\, \lambda^{3N}}; \qquad \lambda \equiv h/(2\pi mkT)^{\frac{1}{2}}. \tag{1-3}$$

Ω is the volume of the system, which is assumed to be of macroscopic dimensions. λ is the mean thermal deBroglie wavelength.

We are concerned only with the effects of the interactions between the particles. Since by assumption the potential energy V does not depend on velocity, the kinetic energy part of Eq. (1-2) can be separated off, so that the partition function has the form

$$Z = Z_0 \frac{1}{\Omega^N} \int d\mathbf{r}_1\, d\mathbf{r}_2 \cdots d\mathbf{r}_N e^{-\beta V}$$

$$\equiv Z_0 \langle \exp(-\beta V) \rangle. \tag{1-4}$$

The expectation value, $\langle f(\mathbf{r}_1, \ldots \mathbf{r}_N) \rangle$, is defined as the average of the function f according to the following probability distribution: The $3N$ variables $\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \ldots, \mathbf{r}_N$ are distributed with a uniform probability density of $(1/\Omega)^N$ inside the box of volume Ω, and zero if any of the $\mathbf{r}_j$ lie outside the box. The "correlation" free energy ΔF, due solely to the interactions between the particles, then assumes the simple form

$$-\beta \Delta F \equiv -\beta(F - F_{\text{ideal}}) = \log(Z/Z_0) = \log \langle \exp(-\beta V) \rangle. \tag{1-5}$$

We further assume that the potential energy V, besides being independent of velocity, is a sum of pair interactions $v_{ij} = v(\mathbf{r}_i - \mathbf{r}_j)$, where $\mathbf{r}_i$ denotes the position of the ith particle:

$$V = \sum_{1 \leqslant i \leqslant j \leqslant N} v_{ij} = \tfrac{1}{2} \sum_{i \neq j} v_{ij}. \tag{1-6}$$

There are $N(N-1)/2$ terms in this sum.

We shall often encounter expressions like the right-hand side of Eq. (1-5). We shall, therefore, recall some results from the theory of probability, in which similar functions appear. For instance, the "moment generating function" $\phi(t)$

$$\phi(t) \equiv \langle \exp(tx) \rangle = \sum_{n=0}^{\infty} \frac{t^n \langle x^n \rangle}{n!} \tag{1-7}$$

generates all the moments of the random variable x according to any probability distribution (indicated by the brackets) for which $\phi(t)$ exists. In the problem at hand, t is $-\beta = -1/kT$, and the random variable x is $V = (1/2) \Sigma v_{ij}$.

The physical quantity of interest is $\log \phi$ instead of ϕ (see Eq. (1-5)). By expanding the exponential, and then the logarithm, in a Taylor series one derives the expansion

$$\log \langle \exp (tx) \rangle = \sum_{n=1}^{\infty} \frac{t^n M_n}{n!} . \tag{1-8}$$

The coefficients M_n have many simple and useful properties which we shall exploit frequently. The M_n are called "semi-invariants," or "cumulants." It is easily checked that the first few semi-invariants are

$$M_1 = \langle x \rangle \tag{1-9}$$

$$M_2 = \langle x^2 \rangle - \langle x \rangle^2 \tag{1-10}$$

$$M_3 = \langle x^3 \rangle - 3\langle x^2 \rangle \langle x \rangle + 2\langle x \rangle^3 \tag{1-11}$$

$$M_4 = \langle x^4 \rangle - 4\langle x^3 \rangle \langle x \rangle - 3\langle x^2 \rangle^2 + 12\langle x^2 \rangle \langle x \rangle^2 - 6\langle x \rangle^4. \tag{1-12}$$

The general expression is

$$M_n = \sum_{\substack{\{n_i\} \\ \left(\sum_i i n_i = n\right)}} (-1)^{\sum_i n_i - 1} \left(\sum_i n_i - 1 \right)! \prod_i \left[\frac{1}{n_i!} \left(\frac{\langle x^i \rangle}{i!} \right)^{n_i} \right]. \tag{1-13}$$

The meaning of the restriction is that one sums over all sets of numbers $\{n_i\}$ satisfying $\sum_i i n_i = n$.

The advantage of working with Eq. (1-8) as opposed to Eq. (1-7) is made clear by considering the two random variables, x and y. In an obvious notation:

$$\log \langle \exp [t(x + y)] \rangle = \sum_n \frac{t^n}{n!} M_n^{(x+y)}. \tag{1-8'}$$

For independent variables x and y, the left-hand side of Eq. (1-8') is also

$$\log \langle \exp (tx) \rangle \langle \exp (ty) \rangle = \log \langle \exp (tx) \rangle + \log \langle \exp (ty) \rangle.$$

Comparison with Eq. (1-8) then yields the important result:

$$M_n^{(x+y)} = M_n^{(x)} + M_n^{(y)}, \tag{1-14}$$

that is, all cross terms involving independent variables vanish in the semi-invariant expansion.

Evidently, if one has the set of independent variables $\{x_i\}$ then

$$M_n^{\left(\sum x_i\right)} = \sum_i M_n^{(x_i)}. \tag{1-14'}$$

As a simple illustration of the cancellation of the cross terms we work out M_2^{x+y}. From Eq. (1-10), for independent x and y

$$\begin{aligned} M_2^{(x+y)} &= \langle (x+y)^2 \rangle - \langle x+y \rangle^2 \\ &= (\langle x^2 \rangle + 2\langle x \rangle \langle y \rangle + \langle y^2 \rangle) - (\langle x \rangle^2 + \langle y \rangle^2 + 2\langle x \rangle \langle y \rangle) \\ &= \langle x^2 \rangle - \langle x \rangle^2 + \langle y^2 \rangle - \langle y \rangle^2 \\ &= M_2^{(x)} + M_2^{(y)}. \end{aligned}$$

We are now prepared to evaluate the free energy. According to Eqs. (1-5) and (1-8) (Brout (1))

$$-\beta \Delta F = \sum_{n=1}^{\infty} \frac{(-\beta)^n}{n!} M_n \tag{1-15}$$

In Eq. (1-15) the free energy is given by an expansion in powers of $1/kT$. We will show how this can be converted to a power series in the density.

First let us examine M_1, which is just the average of the total potential energy:

$$M_1 = \langle V \rangle = \frac{1}{\Omega^N} \int d\mathbf{r}_1 \, d\mathbf{r}_2 \cdots d\mathbf{r}_N \left(\sum_{i<j} v_{ij} \right). \tag{1-16}$$

Obviously, every term in the summation gives the same contribution so that

$$\begin{aligned} M_1 &= \binom{N}{2} \frac{1}{\Omega^N} \int d\mathbf{r}_1 \cdots d\mathbf{r}_N v_{ij} \\ &= \binom{N}{2} \frac{1}{\Omega^2} \int d\mathbf{r}_1 \, d\mathbf{r}_2 v(\mathbf{r}_{12}) \end{aligned} \tag{1-16'}$$

The combinatorial factor $\binom{N}{2} = \dfrac{N(N-1)}{2}$ is the number of terms in the sum in Eq. (1-16). We shall always be interested in the limit of infinite systems; i.e., all expressions are to be evaluated in the limit $N \to \infty$, $\Omega \to \infty$, but with $\rho = N/\Omega$ held fixed at some finite value. This amounts to ignoring surface effects, and for consistency we should keep only the leading term in N. Thus, $N(N-1)/2$ is replaced by $N^2/2$, $\binom{N}{n}$ by $N^n/n!$

At this point we digress to remark that the convergence of the various expansions made, although critical to the subject, is only assumed, not proved. Further, even if these series were proved to

converge, then one would have to decide whether the limit $N \to \infty$ could be taken term by term (as we shall do). This latter procedure is much more restrictive than is mere convergence, in that it demands uniform convergence of the series. Apart from the question of convergence of the expansion in β is the problem posed by certain types of potentials for which the various integrals in the M_n's may be undefined. Examples of this behavior are so common (e.g. hard cores, coulomb potentials) that we cannot ignore this trouble. The usual procedure in such cases is to work out the problem for a similar potential, for which all quantities are well defined. The series can often be rearranged in such a way that no divergences appear when the originally troublesome potential is recovered in the appropriate limit.

For example, intermolecular potentials generally have a repulsive core so that the integrals of v and its powers diverge at small distances. However, the irreducible cluster integrals entering into the Ursell-Mayer expansion are well defined. At intermediate stages of the derivation of the cluster expansion one may use potentials with a large constant finite value for r less than some small value, say $r \leqslant a$. Similarly, for potentials decreasing less rapidly than $1/r^3$ at large r one can supply a convergence factor $e^{-\lambda r}$ and search for an arrangement of terms such that the limit $\lambda \to 0$ exists.

In order to evaluate Eq. (1-16′) we introduce the coordinate transformation

$$\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2; \quad \mathbf{R} = \tfrac{1}{2}(\mathbf{r}_1 + \mathbf{r}_2). \tag{1-17}$$

This transformation has a Jacobian of unity. Then according to the preceding discussion the integral over $\mathbf{R}$ gives a factor Ω, and Eq. (1-16′) becomes

$$\lim_{\substack{N,\Omega\to\infty \\ N/\Omega=\rho}} \left(\frac{M_1}{N}\right) = \tfrac{1}{2}\rho \int v(\mathbf{r})\, d\mathbf{r}. \tag{1-18}$$

It will be noted that $M_1 = \langle V \rangle$ is an extensive quantity, as is the free energy ΔF.

More interesting results are found for M_2:

$$M_2 = \langle V^2 \rangle - \langle V \rangle^2 \tag{1-19}$$

$$M_2 = \sum_{i<j,\,k<l} (\langle v_{ij} v_{kl} \rangle - \langle v_{ij} \rangle \langle v_{kl} \rangle). \tag{1-20}$$

The various terms contributing to Eq. (1-20) may be classified according to the number of subscripts in common.

(*a*) *No indices in common* ("*unlinked*" *terms*). A typical term of this kind is $\langle v_{12}v_{34}\rangle$. Evidently the variables $\mathbf{r}_{12}$ and $\mathbf{r}_{34}$ may be varied independently, so that, according to the previous discussion, v_{12} and v_{34} are independent and give no contribution to M_2.

$$\langle v_{12}v_{34}\rangle = \langle v_{12}\rangle\langle v_{34}\rangle. \tag{1-20'}$$

Thus every unlinked term, arising from $\langle v_{ij}v_{kl}\rangle$ will be cancelled by its counterpart $\langle v_{ij}\rangle\langle v_{kl}\rangle$ in Eq. (1-20).

(*b*) *One index in common* ("*reducibly-linked*" *terms*). An example is $\langle v_{12}v_{23}\rangle$. It is easily seen that for the problem at hand this term also factorizes. This is because of the homogeneity of the medium: we may choose $\mathbf{r}_2$ as origin and integrate over $\mathbf{r}_1$ and $\mathbf{r}_3$. But it does not matter where $\mathbf{r}_2$ is taken to be since the domain of integration is infinite.

$$\langle v_{12}v_{23}\rangle = \frac{1}{\Omega^2}\int d\mathbf{r}_{12}\, d\mathbf{r}_{23} v_{12}v_{23} = \langle v_{12}\rangle\langle v_{23}\rangle \tag{1-20''}$$

This factorization among the reducible parts for the second order reducibly-linked cluster can be extended to any order, as long as this order does not grow with N. Again, factorization implies the vanishing of the reducibly-linked term.

(*c*) *Both pairs of indices in common* ("*irreducibly-linked*" *term*). An example is $\langle v_{12}{}^2\rangle$. In general any term which is neither unlinked nor reducibly linked is said to be irreducible.

Thus M_2 is given by the irreducible terms alone.

$$M_2 = \sum_{i<j} [\langle v_{ij}{}^2\rangle - \langle v_{ij}\rangle^2]. \tag{1-21}$$

Another important point is made evident by comparing the order of magnitude of the two types of terms remaining in M_2.

$$\langle v_{ij}{}^2\rangle = \frac{1}{\Omega}\int v_{ij}{}^2\, d\mathbf{r}_{ij} = O\left(\frac{1}{N}\right),$$

$$\langle v_{ij}\rangle^2 = \left(\frac{1}{\Omega}\int v_{ij}\, d\mathbf{r}_{ij}\right)^2 = O\left(\frac{1}{N^2}\right).$$

Here we have tacitly assumed that the integrals converge. Thus, it is necessary to include only the leading term in Eq. (1-21). As in M_1, there are $N(N-1)/2$ identical contributions so that finally one can write:

$$\lim_{N,\,\Omega\to\infty}\left(\frac{M_2}{N}\right) = \tfrac{1}{2}\rho\int v^2(\mathbf{r})\, d\mathbf{r}. \tag{1-22}$$

It is important to notice that the cancellation of unlinked terms is essential in order that M_2, and hence ΔF, be an extensive quantity (i.e. proportional to N, or Ω). Consider, for example, $\sum_{i<j,k<l} \langle v_{ij}\rangle\langle v_{kl}\rangle$ where $(ij) \neq (kl)$. Each $\langle v_{ij}\rangle$ is of order $(1/\Omega)$. The number of terms contributing to the sum is proportional to $(N^2)^2$. Thus, the entire contribution of the term under consideration is of order N^2. $(N/\Omega = \rho$ is regarded as fixed and finite.) The worst singularity in M_n arising from $\langle V^n\rangle$ is of order N^n. This situation already suggests the theorem,

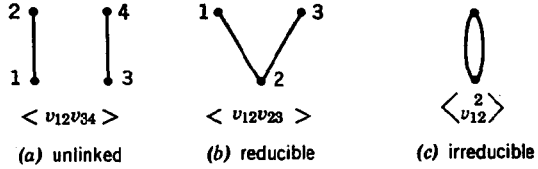

Fig. 1.1. The basic types of contributions to the expectation value $\langle v_{ij}v_{kl}\rangle$ are classified according to the number of overlapping indices.

which we will prove below, that all such terms giving rise to spurious size dependence cancel exactly. This fact actually follows from the theorem expressed in Eq. (1-14), once it is observed that the v products entering into unlinked and reducible parts behave as statistically independent entities.

Before proceeding, it is convenient to introduce a graphical convention for the expectation value $\langle v_{ij}v_{kl}\cdots v_{vs}\rangle$. We shall not need to consider products of expectation values, even though these occur in M_n, because such products always cancel or are $0(1/N)$ relative to the irreducible terms arising from the first term in M_n. The graph corresponding to $\langle v_{ij}v_{kl}\cdots v_{vs}\rangle$ is constructed as follows: for each particle (subscript on v) draw a vertex (point); then draw a bond (line) between the pair of suffixes labeling each v. The topological structure of the resultant geometrical figure depends on how the various particle labels repeat. If the figure contains two or more pieces not joined by any bond then the graph is said to be *unlinked*. If the figure can be separated into two unconnected pieces by cutting at one vertex, the figure is said to be *reducible*, or *reducibly linked*. The remaining graphs are called *irreducible*. Figure 1-1 illustrates these concepts for the graphs arising from $\langle v_{ij}v_{kl}\rangle$ in M_2.

Most of the complexity arising in higher order semi-invariants already occurs in M_3. Therefore, we analyze this term in detail.

$$M_3 = \sum_{\substack{i<j,k<l \\ m<n}} \{\langle v_{ij}v_{kl}v_{mn}\rangle - 3\langle v_{ij}v_{kl}\rangle\langle v_{mn}\rangle + 2\langle v_{ij}\rangle\langle v_{kl}\rangle\langle v_{mn}\rangle\} \quad (1\text{-}23)$$

Consider first those terms in the sum for which the v's have no labels in common (Fig. 2.2*a*). Clearly

$$\langle v_{ij}v_{kl}v_{mn}\rangle = \langle v_{ij}v_{kl}\rangle\langle v_{mn}\rangle = \langle v_{ij}\rangle\langle v_{kl}\rangle\langle v_{mn}\rangle$$

so that by Eq. (1-23) the contribution of terms with no overlapping indices is identically zero. (Note that the size dependence of these terms is given by $(N^2)^3/\Omega^3 \propto N^3$.) There is one other type of unlinked graph coming from $\langle v_{ij}v_{kl}v_{mn}\rangle$, namely that graph in which one pair of the v's has at least one index in common, the remaining v having no index in common with the aforementioned pair (Figs. 2.2*b*) and 2.2*c*). Example: $\langle v_{ij}v_{jk}v_{mn}\rangle$ with $m, n \neq i, j, k$. For this set of indices $\langle v_{ij}v_{jk}v_{mn}\rangle$ is equal to $\langle v_{ij}v_{jk}\rangle\langle v_{mn}\rangle$. For $i \neq k\langle v_{ij}v_{ik}\rangle = \langle v_{ij}\rangle\langle v_{jk}\rangle$ and the argument is same as above. To see that such terms give no contribution for $i = k$ consider the second term in Eq. (1-23) for a fixed set of indices obeying the restriction $m, n \neq i, j$. This same set of numbers can arise in three ways from $\langle v_{ij}v_{kl}v_{mn}\rangle$ depending on which of the three v's takes on the m, n specified in the second terms. Thus, all unlinked graphs arising from $\langle v_{ij}v_{kl}v_{mn}\rangle$ are cancelled off by the remaining terms in M_3. The terms in Figs. (1.2*b*) and 1.2*c*) would give contributions to M_3 proportional to N^2, were it not for the cancellation.

Next, consider the reducible graphs, Fig. 1.2*d* to 1.2*f*. Figure 1.2*d* is of the type $\langle v_{ij}v_{jk}v_{kl}\rangle$, $i \neq k \neq l \neq i$, and factorizes into $\langle v_{ij}\rangle\langle v_{jk}\rangle\langle v_{kl}\rangle$ on passing to relative coordinates, as in Eq. (1-20′). Similarly, Fig. 1.2*e* gives $\langle v_{ij}v_{kj}v_{lj}\rangle = \langle v_{ij}\rangle\langle v_{kj}\rangle\langle v_{lj}\rangle$, $l \neq i \neq k \neq l$. The situation is then completely analogous to Fig. 1.2*a*; cancellation is immediate. The contribution of Fig. 1.2*f* cancels as did Fig. 1.2*c* once it is noticed that $\langle v_{ij}{}^2v_{jk}\rangle = \langle v_{ij}{}^2\rangle\langle v_{jk}\rangle$. Thus only the irreducible graphs (Figs. 1.2*g* and 1.2*h*) remain in M_3. These arise from terms having the structure $\langle v_{ij}{}^3\rangle$ or $\langle v_{ij}v_{jk}v_{ik}\rangle$, respectively. Those terms beyond the first in M_3 that have not been used up to cancel the unlinked and reducible clusters have precisely the same distribution of indices as have the irreducible graphs whose expectation values were indicated above. However, they have at least one more expectation value than the leading term, and are therefore of order $1/N$ times the leading term. Thus, in the limit

$N \to \infty$ only the irreducible clusters arising from the first term give a finite contribution to M_3/N. This fact (true for all orders) explains the importance of the irreducible clusters, and further indicates why it is unnecessary to invent a notation for the products of expectation values.

	Graph	Volume dependence	No. of terms in sum	Order of magnitude of contribution
(a)		Ω^{-3}	N^6	N^3
(b)		Ω^{-3}	N^5	N^2
(c)		Ω^{-2}	N^4	N^2
(d)		Ω^{-3}	N^4	N
(e)		Ω^{-2}	N^3	N
(f)		Ω^{-3}	N^4	N
(g)		Ω^{-1}	N^2	N
(h)		Ω^{-3}	N^4	N

Fig. 1.2. All terms arising from $\langle v_{ij}v_{kl}v_{mn}\rangle$ in the third order semi-invariant are classified according to their order of magnitude.

Our discussion of M_3 is completed by noticing that there are $\left(\frac{N(N-1)}{2}\right)$ identical contributions of the type of Fig. 1.2*g* and $N(N-1)(N-2)$ of the type Fig. 1.2*h*. Thus

$$\lim_{N\to\infty}\left(\frac{M_3}{N}\right) = \frac{\rho}{2}\int v^3(r)\,d\mathbf{r} + \rho^2\int v_{12}v_{23}v_{31}\,d\mathbf{r}_{12}\,d\mathbf{r}_{23} \qquad (1\text{-}24)$$

The important result in our evaluation of M_2 and M_3 is that the unlinked parts drop out (no indices in common) as do the reducible parts (at least one index in common). We should like to rephrase the analysis in slightly different language in order to make evident the generalization to higher order. Whenever unlinked or reducible graphs arise in a semi-invariant, a factorization takes place analogous to