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Abstract. The coupled cluster method (CCM) is nowadays widely recognised as providing one of the most powerful, most universally applicable, and numerically most accurate at attainable levels of implementation, of all available *ab initio* methods of microscopic quantum many-body theory. The number of successful applications of the method to a wide range of physical and chemical systems is impressively large. In almost all such cases the numerical results are either the best or among the best available. A typical example is the electron gas, where the CCM results for the correlation energy agree over the entire metallic density range to within less than one millihartree per electron (or $< 1\%$) with the essentially exact Green's function Monte Carlo results.

What has since become known as the normal (NCCM) version of the method was invented some forty years ago to calculate the ground-state energies of closed-shell atomic nuclei. Extensions of the CCM have since been developed to calculate excited states, energies of open-shell systems, density matrices and hence other properties, sum rules, and the sub-sum-rules that follow from embedding linear response theory within the NCCM. Further extensions deal with the general dynamics of quantum many-body systems, and with their mixed states appropriate, for example, to their behaviour at nonzero temperatures. More recently, a so-called extended (ECCM) version of the method has been introduced. It has the same ability as the NCCM to describe accurately the local properties of quantum many-body systems, but it also has the potential to describe such global phenomena as phase transitions, spontaneous symmetry breaking, states of topological excitation, and nonequilibrium behaviour.

The role of the CCM within modern quantum many-body theory is first surveyed, by a comparison with, and discussion of, the alternative microscopic formulations. We then discuss the method and each of its individual components in considerable detail. Our overall aim is to stress the broad applicability of the method. To that end we introduce and exploit a very general theoretical framework in which to formulate the key ideas and to develop the theory. We end with a brief review of the applications of the method to date.

1 Introduction

1.1 Many-Body Systems

Many-body physics or quantum many-body theory (QMBT) is not the rather specialised subfield of physics that it is sometimes believed to be. On the contrary, we are nearly always faced with many-particle situations at the fundamental level. Thus, we are especially interested in formulations of QMBT

at the fully microscopic or *ab initio* level, which are powerful enough to treat, both in principle and in practice, the full range of many-body and field-theoretic systems. Since these comprise nearly all of the physical world around us, according to our present degree of understanding, the topic of QMBT occupies a key position in modern theoretical physics.

Examples of quantum many-body systems abound. Thus, it is clear that in fields like molecular, solid-state, and nuclear physics most of the fundamental objects of discourse are interacting many-body systems. But even in elementary particle physics one is usually dealing with more than one particle. For example, at some level of reality a nucleon is made up of three quarks interacting via gluons and surrounded by a cloud of mesons, which are themselves composed of quark-antiquark pairs. Even more fundamentally, even the "physical vacuum" of any quantum field theory is endowed with an enormously complex infinite many-body structure due to the virtual excitation of the particles allowed by the theory under discussion. Table 1 gives an incomplete but illustrative set of examples of quantum many-body systems. It is a primary aim of modern QMBT to derive and explore methodologies which are broad enough to be able to be applied to all such systems, and powerful enough to describe their key emergent macroscopic properties from a knowledge only of the underlying microscopic interactions. The coupled cluster method (CCM) under discussion here now plays a central role in modern QMBT since, as we shall explore in detail, it has the potential to fulfill these key criteria amply.

1.2 Many-Body Methods

From our discussion above it should be clear that although techniques to deal with interacting many-body systems are likely to have separately arisen from specific physical contexts, they may also be of more general applicability and interest. The CCM and its birth some 40 years ago within nuclear physics [1,2] is just one example. Nevertheless, if we limit ourselves to comparisons with other fundamental and "universal" tools, the number of available methods is surprisingly small. Before we enumerate them, however, we emphasise that there exist also many other macroscopic or phenomenological or approximate or empirical descriptions and techniques, which have proven to be successful in one or other context. They are, nevertheless, excluded from our present considerations because, by their very nature, they are "specialist" tools which are tailor-made for a particular system or application, and because our emphasis here is on techniques that have a more universal appeal.

The main microscopic techniques fall essentially into seven main classes:

- **Quantum Monte Carlo (QMC) Methods:** These come in several varieties, e.g., Green's function Monte Carlo, diffusion Monte Carlo, path-integral Monte Carlo. Each aims to solve the N-body Schrödinger equation by stochastic simulation, typically for $N \lesssim 1000$, with extrapolation

Table 1. Examples of quantum many-body systems.

System	Particles	Interaction	Infinite Limit
Molecular clusters	e.g., CO_2 molecules	van der Waals type (i.e., repulsive core + attractive tail)	Gases; fluids; molecular crystals
Rare gas atom clusters	e.g., He or Ar atoms	Interatomic potentials (e.g., van der Waals type)	Rare gas; liquid; crystal
Metallic atom clusters	e.g., Na or Ag atoms	Interatomic potentials	Metallic crystal
Atoms and molecules	Electrons and nuclei	Mainly Coulombic (+ relativistic/QED effects)	Infinite Thomas–Fermi atom
Quantum spin clusters	Spins on a spatial lattice	e.g., Heisenberg interaction	Magnetic phases (e.g., antiferromagnets)
Electron clusters	Electrons (and holes) on a regular spatial lattice	e.g., Hubbard model, t–J model	Strongly interacting electron-lattice systems (e.g., high-T_c superconductors)
Nuclei ($A = 2Z$)	(i) α-particles	2-body phenomenological potentials (repulsive core	Alpha matter
	(ii) Nucleons	+ attractive tail); also 3-body potentials needed for accurate fits to experimental data	"Standard" nuclear matter
	(iii) Baryons and mesons	Field-theoretic interaction (sometimes replaced by boson-exchange potentials)	Nuclear matter as "baryon-meson soup"
	(iv) Quarks and gluons	(Hints from) QCD	Quark-gluon plasma
Elementary particles (models from quantum field theory)			
(i) Leptons and exchange bosons		Electroweak interaction (Weinberg–Salam theory)	Already there! (– even for the physical vacuum!)
(ii) Quarks and gluons		(Hints from) QCD	

to the thermodynamic limit, $N \to \infty$, using finite-size scaling or other techniques. The QMC methods typically require either that the many-body wave function is everywhere non-negative (as for bosonic ground states) or that its nodal surface structure is known or well approximated. The latter requirement leads to the infamous "fermion minus sign problem", which still exists as a serious limitation on the practical usefulness of QMC techniques.

- **Time-Independent Perturbation Theory (TIPT):** This method basically provides an expansion in powers of the interaction coupling constant. It finds a diagrammatic representation in terms of so-called Goldstone diagrams. Its main drawback is that one needs in numerical implementations to neglect all terms which are, somehow, deemed to be unimportant or which, more usually, simply cannot be handled in practice.
- **Green's Function (GF) or Propagator Methods:** These methods are basically a reformulation of time-dependent perturbation theory and, as such, can be given a diagrammatic representation in terms of so-called Feynman diagrams. They suffer similar drawbacks to those alluded to above in the case of TIPT. GF methods are formulated as time-dependent (i.e., dynamic) equations for matrix elements (or propagators) which describe the propagation of clusters of particles in the many-body medium.
- **Variational Methods:** The aim of all variational calculations is to compute the energy expectation value, $\langle H \rangle$, with a trial correlated many-body wave function, typical forms of which are the Jastrow or Jastrow–Feenberg varieties. Since $\langle H \rangle$ cannot usually be calculated in closed form, various techniques are used. Some typical ones are Monte Carlo evaluation of the implicit multi-dimensional integrals, various cluster expansions, and various partially re-summed cluster sub-series (e.g., the hypernetted chain approximation).
- **Correlated Basis Function (CBF) Method:** Variational calculations suffer from the main drawback that even if $\langle H \rangle$ is calculated exactly for a given trial wave function, one has only an upper bound on the exact energy and an approximation to the exact many-body wave function. CBF techniques aim to remove this restriction by providing a scheme in which to improve systematically on a trial wave function (typically of Jastrow or similar form) by introducing a *complete* set of correlated basis functions. CBF techniques typically enable the short-range correlations which are important for many physical applications to be efficiently incorporated from the outset.
- **Configuration-Interaction Method (CIM)** (or generalised many-body shell model): This method typically aims to diagonalise the Hamiltonian in a finite subspace of the full many-body Hilbert (or Fock) space. Despite its underlying simplicity, it suffers from the severe drawback for

many-body applications of not satisfying the important requirements of size-extensivity and size-consistency.

- **Coupled Cluster Method (CCM):** By utilising an exponentially correlated form of the wave function, the CCM cures the size-extensivity problem inherent in the previous CIM. The CCM has the nice feature that if its basic equations are iterated one regains TIPT and the associated Goldstone diagrams. Nevertheless, one need not, and usually does not, solve the equations this way, thereby circumventing the main drawback of TIPT methods. The characteristic exponential form of the CCM wave functions correctly counts independent clusters excitations from a suitably chosen model or reference state, which itself can take a variety of forms depending on the system and on its particular phase under study.

The QMC, variational, and CBF methods are all extensively discussed by other contributors to this volume. It is nevertheless worthwhile to spend a little while in discussing the salient features of all of the methods (apart from QMC techniques), in order to put them properly in context before turning to a detailed discussion of the CCM.

Both the TIPT and GF methods are perturbative in origin. In almost all cases of interest various infinite partial resummations are performed out of necessity, either because the series diverges badly or is otherwise ill-defined from the outset. Well-known examples include the ladder-diagram summation (for hard-core or other strongly repulsive short-range potentials) which leads to the Bethe–Brueckner–Goldstone G-matrix expansion or the hole-line expansion (and see, e.g., Refs. [3–5]); and the bubble- or ring-diagram summation of the random phase approximation (RPA) [6,7] (for long-range potentials of the Coulomb type). For accurate numerical calculations of most realistic many-body systems it is usually found to be necessary to incorporate at least the self-consistent sum of all ring and ladder diagrams.

Even in such rearranged or partially resummed examples, one is still ultimately forced in practice to neglect infinite classes of terms that are presumed to be unimportant or that cannot easily be incorporated into a higher approximation. Furthermore, it is usually difficult in realistic applications to justify the retention of certain terms at the expense of neglecting others. The history of microscopic nuclear theory, for example, is littered with incorrect or misleading calculations which amply illustrate the danger of the blind inclusion of extra diagrams, as has been forcefully pointed out many times by Kümmel and his collaborators [8,9]. In quantum chemistry, the TIPT techniques provide the usual means of doing many-body perturbation theory (MBPT), or what has increasingly become known in chemistry as Møller–Plesset perturbation theory. In chemical applications it is usual to include all terms through some given finite order in the electron interactions. Full fourth-order, MBPT(4), calculations are now commonplace, for example [10].

Various propagator or GF techniques are also still widely used in quantum chemistry (and see, e.g., Refs. [11,12]), where they are particularly used for molecular spectroscopic properties, as well as in other areas. Nevertheless, on the wider front, GF methods suffer from many of the same drawbacks as the TIPT techniques discussed above. Thus, with one important potential caveat, neither TIPT nor GF methods are nowadays generally considered within quantum many-body theory to be sufficiently accurate or sufficiently versatile to be candidates for the position of a universal, high-precision method. The only real exception to this viewpoint is provided by the so-called *parquet diagram,* or *planar theory,* approach within the broader GF scheme.

The ideas behind parquet theory were introduced [13] at around the same time that Coester first introduced the CCM. The specific context for their introduction was certain problems in particle physics. Similar equations were later used to study X-ray absorption and emission in metals [14]. However, it was not until 1979 that the potential of parquet theory for the study of strongly interacting condensed Bose systems was noted by Ripka [15]. He called attention to the fact that the hypernetted chain (HNC) equations of the Jastrow variational approach shared several very desirable features with the parquet equations. At the time the HNC approximation to the variational approach, which is discussed in detail in the contribution to this volume by Fantoni and Fabrocini, was meeting with considerable success in dealing with such extended bosonic systems as liquid ^{4}He. Ripka noted that just as the HNC equations of variational theory treated both the long- and short-range correlations simultaneously and consistently, so too would the parquet equations of perturbation theory, if only they could be solved.

This astute observation of Ripka provided the stimulus for the 1982 study of Jackson, Lande, and Smith [16] which examined the parquet equations for bosons in great detail. They also showed how these otherwise rather formidable equations could be rendered tractable, and hence solved numerically, by the introduction of simple localising approximations.

The basic concepts of parquet theory are both simple and appealing. The method first focusses on the effective two-body interaction, which it then expresses in terms of a large and physically interesting class of Feynman diagrams. These so-called parquet diagrams are a particular self-consistent sum of ring, ladder, and vertex correction terms for the two-body Green's function. Most important physically is the fact that the two-body (i.e., particle–particle and hole–hole) ladder diagrams and the particle–hole ring diagrams are iterated together *in a maximal fashion.* Although the full two-body parquet equations are highly complex, their local counterparts were applied to such model bosonic systems as liquid ^{4}He using the Lennard–Jones potential, and neutron matter treated as a Bose system interacting via the Reid soft-core $^1S_0$ interaction. Not surprisingly in view of Ripka's comments, good agreement was found with the corresponding optimised Jastrow HNC results.

At the formal level it was later demonstrated [17] that a similar, but nonetheless distinct, local form of the full two-body parquet equations is *identical* to optimised Jastrow HNC theory. This result was particularly important since it provided a bridge between the otherwise very disjoint perturbative approaches of TIPT and GF theory on the one hand, and variational approaches on the other. Later formal developments have included a possible extension to fermionic systems [18,19], the inclusion of three-body terms [20], and parquet perturbation theory [21] for bosons, as an expansion in the difference between the exact and approximate propagators, in order to improve systematically upon the local parquet equations. Interesting connections between parquet theory and Baym–Kadanoff theory [22] have also been made [23]. This latter approach is noteworthy in that it uses an initial approximation for the two-body vertex to construct from it a *conserving* vertex, namely one which conserves particle number, momentum, and energy. Despite what is now a fairly large corpus of *formal* developments, the parquet method has not yet been widely applied and tested. Nevertheless, it clearly deserves to be, and it must be considered as a potential candidate for the position of a universal tool in quantum many-body theory. We have stressed it here for just these reasons.

We turn next to the use of variational methods in quantum many-body theory. Perhaps the simplest of these approaches is based on a trial wave function of the (Bijl–Dingle–)Jastrow type [24]. The early calculations of this sort relied on various cluster expansions of the ensuing approximate matrix elements [24–26]. Later on it was realised that the variational approaches may themselves be formulated diagrammatically [15,27,28]. This feature has been of considerable help in constructing such powerful approximations as the Percus–Yevick and HNC summations (which have their origins in the classical theory of liquids) and their variants, both for bosons and fermions [15,29–31]. Various articles and reviews concerning the use of variational methods in quantum many-body theory exist (and see, e.g., Refs. [15,32–35]). The 1979 article by Clark [32] is particularly recommended as what is perhaps still the standard review of the variational theory of extended nuclear matter. Similar reviews (and see, e.g., Ref. [36]) also exist for applications to few-body systems. Finally, Fantoni and other contributors to this volume describe the variational techniques and their applications in some depth, from both a modern perspective and a pedagogical point of view.

The variational approaches sketched above suffer from two fundamental flaws. In the first place the various partial summations of the graphs considered by such approximation schemes as HNC give methods which lose one of the most attractive features of variational formulations, namely that they yield estimates for the energy which are upper bounds to the exact ground-state energy. Secondly, even a complete summation of graphs (or a variational Monte Carlo evaluation of the corresponding expectation values) for a given trial wave function, say of the Jastrow type, gives only the exact

variational result and not the true ground state. This second deficiency may be removed in principle by including more general state-dependent correlations and higher-order correlation functions of the Feenberg type [37]. Alternatively, and more generally, one may extend the Jastrow wave function to a complete set of correlated basis functions (CBF). The CBF method was introduced nearly forty years ago by Feenberg and his collaborators [29,38], and has since been developed largely by Clark and his co-workers [32,39–45]. A very brief and qualitative survey of the method, which contains many more citations of the original CBF literature, has also been given by the present author [46]. Despite the fact that the method and its applications are also extensively discussed in the present volume by Fantoni, Fabrocini, and Krotscheck, it is useful for us also to review the key features of the method for purposes of later comparison with the CCM.

The central ingredient of the CBF method is the direct incorporation of the most important interparticle correlations which are believed to characterise the system under consideration, into the approximate wave functions on which the microscopic description is based. At its simplest level the method considers only a single configuration and hence reduces to ordinary variational theory. As we have seen above, the latter further simplifies to Jastrow theory if the simplest reasonable choice of correlation operator is made.

At its most general level, the CBF method constructs for the N-body system a multiconfigurational correlated basis $\{|\Psi_I\rangle\}$ of normalised but generally nonorthogonal state vectors,

$$|\Psi_I\rangle = \frac{F|\Phi_I\rangle}{\langle\Phi_I|F^\dagger F|\Phi_I\rangle^{\frac{1}{2}}} \quad , \tag{1}$$

in terms of a correlation operator F applied to a complete orthonormal basis $\{|\Phi_I\rangle\}$ of model states. The latter usually carry the correct quantum statistics and any essential symmetries of the system. They would hence provide an adequate lowest-order description of the system if it were not strongly interacting. For an application to molecules, for example, the states $\{|\Phi_I\rangle\}$ would probably be chosen as a set of Slater determinants of some appropriate single-electron orbitals.

The correlation operator F is thus symmetric in the complete sets of single-particle quantum numbers used to denote the N particles. Most importantly, F also possesses the cluster decomposition property, namely that upon separating one subgroup of particles (say 1,2,...,n) far from the rest $(n+1, n+2, ..., N)$ in real space, the operator $F(1, 2, ..., N)$ decomposes into a product,

$$F(1, ..., N) \rightarrow F^{(n)}(1, ..., n)F^{(N-n)}(n+1, ..., N) \quad . \tag{2}$$

It is this property which allows the natural definition of correlation operators $F^{(n)}(1, ..., n)$ for n-body subsystems with $1 \leq n \leq N$, in terms of a given

N-body operator $F \equiv F^{(N)}$. It is also the key ingredient in allowing the derivation of linked cluster expansions for physical quantities.

The simplest choice for F is the state-independent Jastrow form, $F \rightarrow F_J$, specified in its usual coordinate-space form as follows,

$$F_J = \prod_{1 \leq i < j \leq N} f(r_{ij}) \ , \tag{3}$$

where r_{ij} is the relative coordinate for particles i and j. The more general Feenberg form, $F \rightarrow F_F$, is given in terms of higher-order correlation functions as,

$$F_F = \prod_{i<j} f_2(r_{ij}) \prod_{i<j<k} f_3(r_{ij}, r_{ik}, r_{jk}) \cdots \ . \tag{4}$$

Whatever choice for F is adopted, the CBF method devolves onto the computation of the matrix elements H_{IJ} and N_{IJ} of the Hamiltonian H and the unit operator, respectively,

$$H_{IJ} \equiv \langle \Phi_I | F^\dagger H F | \Phi_J \rangle \ ; \quad N_{IJ} \equiv \langle \Phi_I | F^\dagger F | \Phi_J \rangle \ . \tag{5}$$

One assumes that any exact stationary energy eigenstate $|\Xi\rangle, H|\Xi\rangle = E|\Xi\rangle$, may now be (approximately) expanded in the multiconfigurational basis,

$$|\Xi\rangle = \sum_J c_J |\Psi_J\rangle \ . \tag{6}$$

The Schrödinger equation is then decomposed as usual into the coupled set of linear generalised eigenvalue equations,

$$\sum_J (H_{IJ} - E N_{IJ}) c_J = 0 \ , \tag{7}$$

which have a nontrivial solution for the coefficients $\{c_J\}$ if and only if E satisfies the secular equation,

$$\det(H_{IJ} - E N_{IJ}) = 0 \ . \tag{8}$$

Clearly, the accurate numerical evaluation of the matrix elements $\{H_{IJ}\}$ and $\{N_{IJ}\}$ is far from trivial. In this context various cluster-expansion techniques have been developed [26,47], which themselves also provided a basis for much of the later diagrammatic analysis of both diagonal elements (expectation values) [27–31] and off-diagonal elements [40]. This latter work led to the definition of the CBF effective interaction, and to many illuminating connections with conventional diagrammatic MBPT. Exploration of these connections has allowed techniques from standard TIPT as developed for weakly-interacting systems to be taken over into their CBF counterparts for application to strongly-interacting systems (and see, e.g., Ref. [44]).

Finally, however the matrix elements $\{H_{IJ}\}$ and $\{N_{IJ}\}$ are derived, the remaining generalised eigenvalue problem of (7) and (8) must then be attacked in a systematic, approximate, or heuristic manner. Various such methods have been developed within the general CBF framework. These include nonorthogonal perturbation theory [39]; straightforward CIM-type diagonalisation in the space spanned by some subset of the full correlated basis [39,48]; and a correlated version of the usual RPA [6], which now performs a semiclassical treatment of small-amplitude oscillations about an equilibrium correlated configuration [41,42,44,45]. There have even been attempts to formulate a generalised version of coupled cluster theory in the CBF basis [49].

We turn finally to the CIM [50] and the CCM, where the latter is our main concern. For present overview purposes we merely remind the reader that the CIM is basically a straightforward diagonalisation of the Hamiltonian in some multiconfigurational subspace of the full many-body Hilbert space. One of its main advantages is that it is also easily and equivalently formulated variationally, in a parametrisation that preserves the manifest Hermitian adjoint relationship between corresponding bra and ket wave functions. In this way we see that each of the set of approximate energy eigenvalues so obtained provides an upper bound to the corresponding member of the similar set of exact lowest eigenvalues. Furthermore, by the well-known interlacing (or Hylleraas–Undheim) theorem [51], an increase in the size of the multiconfigurational CIM subspace is guaranteed to lead to improved estimates, i.e., to ones which are lower (or, in the worst case, unchanged) in energy.

This advantage of the CIM (which is *not* shared by the CCM) is, however, offset by the disadvantage that the method is not size-extensive [52]. Thus, in diagrammatic language, the CIM contains terms which are not linked (connected).[1] For extended systems this deficiency is fatal. In order to guarantee the size-extensitivity of the energy of the many-body system, it is clearly necessary that the effective Hamiltonian is separable over the various possible subsystems which arise in the corresponding dissociation limits where they are asymptotically separated to large relative distances so that the interactions between them become vanishingly small. This will certainly be the case if the wave operator is multiplicatively separable. The importance of exactly maintaining the separability of the wave function *at any subsequent level of approximation* has been stressed by Primas [53], although its importance for energy calculations was perhaps first realised by Brueckner [54] when using nondegenerate Rayleigh–Schrödinger MBPT to calculate the ground-state energy of infinite nuclear matter. An obvious method of ensuring that the wave operator is multiplicatively separable is to write it as the exponential of the so-called cluster operator, which in turn is additively separable and hence

[1] In the present article we use the terms "linked" and "connected" as synonyms; the interested reader should beware, however, that in the context of open-shell versions of the CCM it is usual to distinguish between the two terms.

representable by a sum of connected diagrams. This is the central feature of the original CCM, as described more fully in Sect. 3.

From our discussion to date it should be apparent that the only existing formulations of quantum many-body theory which satisfy *all* of the desirable criteria of being:

- fully microscopic;
- widely applicable to a broad spectrum of both finite and extended systems of interest in physics (and quantum chemistry);
- equally applicable to systems in a spatial continuum or on a discrete (regular) spatial lattice;
- capable of systematic improvement via increasingly higher-order implementations of some well-defined scheme of hierarchical approximations;
- very accurate in practice, i.e., better than or comparable to the highest practicable precisions attainable by any alternative technique, however specialised to the system under discussion; and
- very widely and deeply tested,

and which can hence claim to be universal, high-precision tools, are the CBF and CCM formalisms. Parquet (or planar) theory may also potentially fall into this category, but it has not yet been sufficiently widely tested to be able to judge properly. Naturally, QMC methods also yield excellent results which, when the methods can be implemented, can often be viewed as exact benchmarks, as other contributions to this volume make clear. Nevertheless, it is still often convenient to view QMC techniques as providing a third paradigm in physics, sitting between experiment and microscopic (analytic or semi-analytic) theory.

We note that the CBF method, and the variational techniques which it subsumes and extends, have been widely applied to systems as diverse as finite nuclei [41,45,48]; nuclear matter [32,55–57]; neutron stars [58]; bulk liquid ^{4}He [55,59,60]; unpolarised [55,61] and polarised [44] bulk liquid ^{3}He; the electron gas [61]; various species of electron-spin-aligned bulk atomic deuterium [43]; the lattice Hubbard model of strongly interacting electrons [62]; electronic correlations in atoms [63]; Ising spins in a transverse magnetic field [64]; and $U(1)_3$ lattice gauge field theory in the pure gauge sector [65]. Other contributors to this volume discuss these and other applications in much more detail.

The CCM has been at least as widely and at least as successfully (and accurately) applied in both physics and chemistry as the CBF method, as we shall outline below in Sect. 4.

1.3 Relationships Between Methods

Before turning our attention to the CCM we end the present overview of many-body techniques by remarking that the fact that there exists only a

small number of fundamental *ab initio* methods is not necessarily something to be regretted. Indeed, it is widely believed by quantum many-body theorists that what is much more important nowadays than the development of further techniques is the exploration of the interconnections between existing ones. This is clearly a reflection on the power and universality of the available methods. In this regard we note that there have already been several important confrontations and proposed marriages between existing methods, and we conclude by mentioning some of them.

In the first place, one of the earliest confrontations was provided by the so-called "crisis in nuclear matter theory" which occupied a central place in quantum many-body theory in the 1970s. Two fundamental issues were involved. Thus, there was a disagreement between the lowest-order Brueckner theory calculations of TIPT and various variational calculations, when both were performed with the same internucleon potential. Furthermore, neither method gave good saturation properties when the best available potentials were used. That crisis has now been essentially resolved (and see, e.g., Ref. [37]) to the mutual advantage of both methods. In particular, the later development of Brueckner theory that was necessary for the resolution has greatly added to our understanding of many-body theory and many-body systems. Another very beneficial outcome of this early confrontation was that the power of the variational and the CBF techniques was thereafter much more widely appreciated.

Secondly, we have already noted the important formal role played by parquet theory [17] in demonstrating the exact equivalence between the optimised hypernetted chain (HNC) approximation to the variational Jastrow theory and a particular (approximate) localised form of itself. This result provided a real insight into the relationships between perturbative expansions on the one hand and variational cluster expansions on the other. It also provided a bridge between the two methods, as well as a common language in which to interpret both their features and the results obtained from them.

Thirdly, the relationship of the CCM to TIPT as providing at given levels of truncation (e.g., in the SUBn scheme discussed in detail in Sect. 3) very sophisticated and intricate resummations of various physically important and infinite classes of Goldstone diagrams is by now well known to the practitioners, and will be fully explored in Sect. 3. In particular, the very natural grouping together within the CCM of vast classes of Goldstone diagrams has shown graphically how cautious one must be when using perturbation theory in order "not to split small quantities into large pieces" [66].

A fourth result concerns the relationship of the CIM to the CCM. In particular, the CIM together with the two main modern complete formulations of the CCM which we describe more fully in Sect. 3, and which are known respectively as the normal (NCCM) and extended (ECCM) versions [67], have been shown [68,69] to form a natural closed hierarchy of three increasingly sophisticated parametrisations in which the underlying amplitudes have in-

creasing degrees of connectedness. In the same spirit, as we discuss in Sect. 3, each of these three *independent cluster methods* has also been completely reformulated [67–69] via a variational principle, thereby providing further bridges between perturbative and variational techniques.

Fifthly, and at the level of possible marriages between various of the methods, we have already mentioned the "correlated coupled cluster theory" [49], which was motivated by a desire to combine the best features of both the CCM and CBF approaches. It is unfortunate that, to the best of our knowledge, this proposed marriage has not yet really been put to the test of a real application.

More recently, however, two other such extensions of the CCM have been discussed. Thus, sixthly, in a recent series of papers [70–74] the CCM (and the CIM) have been the subjects of intense investigation in order to incorporate exactly the translational invariance property, which is vital for the accurate treatment of such light nuclei as the alpha-particle. As a consequence, the emphasis has been to formulate the CCM and the underlying correlations directly in coordinate space, rather than in the more usual multiconfigurational Fock-space representation discussed at length in Sects. 2 and 3 below. In this way contact has been made both with more traditional generalised (many-body) nuclear shell-model calculations of the CIM type, with the result that the number of independent configurations can be dramatically reduced [70,71], and also with variational approaches [72,73]. This work now holds out the possibility of combining some of the best elements of the CIM, CCM, and variational approaches. We also note in passing that similar functional forms of the CCM parametrisation of many-body wave functions, rather than the more standard Fock-space or operatorial forms, have also been used recently in the context of lattice field theory. To date, specific applications of the CCM have been made to both discrete and continuous lattice gauge field theories. The former includes the Z(2) case [75], while the latter include both the Abelian U(1) case [75–80] and the non-Abelian SU(2) case [77,81]. Very recent applications of the CCM have also been made to a latticised O(4) nonlinear sigma model of chiral meson field theory [82], wherein the operatorial and functional forms of the CCM have been compared in considerable depth.

Finally, and as a seventh example, recent work [83,84] has been connected with the incorporation of Jastrow-type correlations into the higher-order cluster terms that are otherwise neglected entirely in the standard (e.g., SUBn) CCM truncation schemes discussed in Sect. 3. Indeed, much current effort is being expended in the general direction of inventing new approximation schemes for existing methods. One promising way to do this is by attempting to fuse the best features of different methods.

All of the above various partial mergers hold considerable promise for the future of quantum many-body theory. The interested reader is referred to Ref. [84] for a more detailed, yet still pedagogical, confrontation between the different microscopic techniques discussed above. This paper discusses in

some depth the various ways that two-body correlations in quantum many-body systems are described within different microscopic theories. A particular application of each method is made to the exactly integrable (by the Bethe ansatz [85] method) Lieb model [86,87] of many bosons interacting in one dimension via repulsive pairwise delta-function potentials.

In the remainder of this article we now focus our attention on the CCM. However, before describing the method in Sect. 3, we first discuss the general question of the choice of a suitable reference state (or "generalised vacuum" state) for a given quantum many-body system, with respect to which the dynamic (multiparticle) correlations may be quantitatively described.

2 The Construction of Many-Body Reference States

For many purposes it is extremely convenient to distinguish some *model space*, $\mathcal{M}$, which is a subspace of the full many-body Hilbert or Fock space, $\mathcal{G}$, in which the quantum many-body or quantum field-theoretical problem under discussion is defined. Of particular interest is the case where the subspace is spanned by a suitably chosen set of $(D+1)$ normalised *reference states* $\{|\Phi_i\rangle;\ i=0,1,2,\cdots,D\}$, with $\langle\Phi_i|\Phi_i\rangle=1$. We define two operators, P and Q, as follows,

$$P \equiv \sum_{i=0}^{D} |\Phi_i\rangle\langle\Phi_i| \ ; \quad Q \equiv \mathbb{1} - P \ , \tag{9}$$

where $\mathbb{1}$ is the identity operator in $\mathcal{G}$. If the reference states are orthonormalised,

$$\langle\Phi_i|\Phi_j\rangle = \delta_{ij} \ , \tag{10}$$

the operators P and Q are simply projection operators into and out of the model space, $\mathcal{M}$,

$$P^2 = P \ ; \quad Q^2 = Q \ ; \quad PQ = QP = 0 \ , \tag{11}$$

and we can decompose an arbitrary state $|\Psi\rangle \in \mathcal{G}$ as,

$$|\Psi\rangle = P|\Psi\rangle + Q|\Psi\rangle \ . \tag{12}$$

Although the above projection simply represents an expansion of a wave function in some (suitably chosen and suitably truncated) complete orthonormal set, if, in some sense to be defined below, the states $\{|\Phi_i\rangle;\ i=0,1,\cdots,D\}$ are "zeroth-order" or "starting" or "reference" vectors, then it can be very useful to make this explicit. Furthermore, and more generally, in order to describe many-body correlations quantitatively, we *always* need some appropriate state with respect to which the correlations are defined. Henceforth, we shall call the states $\{|\Phi_i\rangle;\ i=0,1,\cdots,D\}$ the *reference states* or *model states*, and the $(D+1)$-dimensional space $\mathcal{M}$ spanned by them the *reference*

space or *model space*. Later, we shall see that the case $D = 0$ is appropriate to the cases of nondegenerate perturbation theory and to the so-called *single-reference version of the CCM*, while the case $D > 0$ is more appropriate to the cases of degenerate perturbation theory and to the so-called *multi-reference version of the CCM*.

For the moment, we restrict ourselves to the case $D = 0$ of a single model state $|\Phi_0\rangle$. For many purposes, and as we shall see specifically in Sect. 3 for the CCM, it is very useful or even vital for the model state $|\Phi_0\rangle$ to be a *generalised vacuum state*, with respect to which all of the states in $\mathcal{G}$ can be expressed in terms of many-body, creation correlation operators acting on $|\Phi_0\rangle$. Implicit in the above is the enormous simplification that arises if the algebra of all operators in $\mathcal{G}$ and its adjoint space $\mathcal{G}^*$ is spanned by the *two Abelian subalgebras* of (many-body) *multiconfigurational creation operators* $\{C_I^\dagger\}$ and their Hermitian-adjoint counterparts, namely the *multiconfigurational destruction operators* $\{C_I\}$. Both sets of operators are defined with respect to the given model state $|\Phi_0\rangle$. We note that implicit in the compact notation is the fact that the index I is in general a *set-index*, comprising a set of single-particle labels (in some suitable single-particle basis) which completely characterises a given many-body *configuration* in this basis. More specifically, the single-particle labels in the set-index I usually comprise only those needed to describe the states which differ from those occupied in the model state $|\Phi_0\rangle$. For this reason it is convenient to introduce the notation,

$$C_0^\dagger \equiv \mathbb{1} \equiv C_0 \quad . \tag{13}$$

In summary, we require from our sets $\{|\Phi_0\rangle; C_I^\dagger\}$ the following properties,

$$[C_I^\dagger, C_J^\dagger] = 0 = [C_I, C_J] \quad , \tag{14}$$

$$C_I|\Phi_0\rangle = 0 = \langle\Phi_0|C_I^\dagger \quad ; \quad \forall\, I \neq 0 \quad . \tag{15}$$

We further require that the two subalgebras and the state $|\Phi_0\rangle$ are *cyclic* in the following sense,

$$|\Psi\rangle = \sum_I \psi_I C_I^\dagger |\Phi_0\rangle \quad ; \quad \forall\, |\Psi\rangle \in \mathcal{G} \quad , \tag{16a}$$

$$\langle\tilde{\Psi}| = \sum_I \tilde{\psi}_I \langle\Phi_0|C_I \quad ; \quad \forall\, \langle\tilde{\Psi}| \in \mathcal{G}^* \quad , \tag{16b}$$

in terms of some sets of c-number expansion coefficients $\{\psi_I\}$ and $\{\tilde{\psi}_I\}$. The configuration-label space $\mathcal{I} \equiv \{I\}$ must thus be complete (for a given generalised vacuum or model state $|\Phi_0\rangle$) with respect to the many-body configurations. In general, the operators $\{C_I^\dagger\}$ will be products (or sums of products) of suitable single-particle operators. Although it is not vital, it is also convenient to impose the *orthonormality condition*,

$$\langle\Phi_0|C_I C_J^\dagger|\Phi_0\rangle = \delta_{IJ} \quad , \tag{17}$$

on the operators, where δ_{IJ} is a Kronecker delta symbol implying equality between the sets of single-particle (s.p.) labels I and J, i.e., equality under at least one permutation. Henceforth we assume that (17) holds, and hence we have the completeness relation in $\mathcal{G}$,

$$\sum_I C_I^\dagger |\Phi_0\rangle\langle\Phi_0| C_I = \mathbb{1} = |\Phi_0\rangle\langle\Phi_0| + \sum_I{}' C_I^\dagger |\Phi_0\rangle\langle\Phi_0| C_I \quad , \tag{18}$$

where, here and henceforth, a prime on a sum over configuration labels I excludes the term $I = 0$.

We note that it is not, *a priori*, obvious that sets $\{|\Phi_0\rangle; C_I^\dagger\}$ exist with the above properties (14)–(18). We show below, by specific examples to various broad classes of important many-body systems, that not only can such sets generally be found rather easily, but that they are often non-unique in the sense that several sets can be found for use with the same space $\mathcal{G}$.

2.1 Field-Theoretic (Number-Nonconserving) Bose System

An obvious choice for $|\Phi_0\rangle$ in this case is the bare vacuum state, $|0\rangle$; and the multiconfigurational creation operators, $C_I^\dagger$, then become suitably normalised products of single-boson creation operators, $b_\alpha^\dagger$, in some particular single-particle basis,

$$|\alpha\rangle \equiv b_\alpha^\dagger |0\rangle \quad . \tag{19}$$

The single-boson creation and destruction operators, $b_\alpha^\dagger$ and b_α, respectively, obey the usual bosonic commutation relations,

$$[b_\alpha, b_\beta] = 0 = [b_\alpha^\dagger, b_\beta^\dagger] \quad ; \quad [b_\alpha, b_\beta^\dagger] = \delta_{\alpha\beta}\mathbb{1} \quad . \tag{20}$$

In the symmetric boson Fock space $\mathcal{B}$ we have the fundamental completeness relation,

$$\mathbb{1} = |0\rangle\langle 0| + \sum_{n=1}^{\infty} \frac{1}{n!} \sum_{\{\alpha_i\}} b_{\alpha_1}^\dagger \cdots b_{\alpha_n}^\dagger |0\rangle\langle 0| b_{\alpha_n} \cdots b_{\alpha_1} \quad , \tag{21a}$$

as a specific example of the relation (18). However, in (21a) any s.p. index in the products of operators may be repeated any number of times. Hence, it is often more convenient to re-label the s.p. indices as an *ordered* set $\{\bar{\alpha}_i\}$, e.g., $\bar{\alpha}_1 < \bar{\alpha}_2 < \bar{\alpha}_3 \cdots$, such that all members of this set are distinct. The completeness relation (21a) may then be conveniently rewritten in the equivalent form,

$$\mathbb{1} = |0\rangle\langle 0| + \sum_{n=1}^{\infty} \sum_{\{\bar{\alpha}_i\}} \sum_{\{m_i\}} \frac{1}{m_1! \cdots m_k!} \times \left(b_{\bar{\alpha}_1}^\dagger\right)^{m_1} \cdots \left(b_{\bar{\alpha}_k}^\dagger\right)^{m_k} |0\rangle\langle 0| \left(b_{\bar{\alpha}_k}\right)^{m_k} \cdots \left(b_{\bar{\alpha}_1}\right)^{m_1} \quad , \tag{21b}$$

where $\sum_{i=1}^{k} m_i = n$. Thus, in this example, the set-index I becomes the set of occupation numbers,

$$I \rightarrow (m^I_{\tilde{\alpha}_1}, m^I_{\tilde{\alpha}_2}, \cdots) \quad ; \quad m^I_{\tilde{\alpha}_i} = 0, 1, 2, \cdots \quad , \tag{22}$$

and the generic multiconfigurational creation operators $C^\dagger_I$ become

$$C^\dagger_I \rightarrow \prod_i \left[(m^I_{\tilde{\alpha}_i}!)^{-\frac{1}{2}} \left(b^\dagger_{\tilde{\alpha}_i} \right)^{m^I_{\tilde{\alpha}_i}} \right] \quad , \tag{23}$$

where the superscript I in the occupation numbers, $m^I_{\tilde{\alpha}_i}$, reminds us of the particular configuration to which they refer. In the coordinate-space basis, $|\mathbf{r}\rangle = b^\dagger(\mathbf{r})|0\rangle$, for example, we have

$$I \rightarrow (\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_n) \quad ; \quad n = 0, 1, 2, \cdots \quad , \tag{24}$$

and

$$C^\dagger_I \longrightarrow (n!)^{-\frac{1}{2}} \prod_{i=1}^{n} b^\dagger(\mathbf{r}_i) \quad , \tag{25}$$

in terms of the creation field operator, $b^\dagger(\mathbf{r})$.

We note that for the special case of *single-mode bosonic field theory*, in which only one mode exists, with a corresponding creation operator $b^\dagger$, we have

$$I \longrightarrow n \quad ; \quad n = 0, 1, 2, \cdots \quad , \tag{26}$$

and

$$C^\dagger_I \longrightarrow (n!)^{-\frac{1}{2}} (b^\dagger)^n \quad . \tag{27}$$

Such single-mode bosonic field theories are useful nontrivial examples of real quantum field theories in $(d+1)$ dimensions, where d is the number of spatial dimensions, for the limiting case $d = 0$. Clearly, such $(0+1)$-dimensional theories also map precisely onto one-body quantum mechanics through the usual mapping of the single-mode creation and destruction operators, $b^\dagger$ and b, onto the usual position and momentum operators, $\hat{x}$ and $\hat{p}$, respectively,

$$\hat{x} = 2^{-\frac{1}{2}} (b^\dagger + b) \quad ; \quad \hat{p} = 2^{-\frac{1}{2}} \mathrm{i} (b^\dagger - b) \quad . \tag{28}$$

2.2 Number-Conserving Bose System

We now consider a system of N bosons in a box of volume Ω, appropriate to a system such as liquid ^{4}He. We shall usually be interested in such systems in the *thermodynamic limit*, where $N \rightarrow \infty$ and $\Omega \rightarrow \infty$, such that the density, $\rho \equiv N/\Omega$, remains constant. In such cases an obvious candidate for the model state $|\Phi_0\rangle$ in the fixed-N symmetric boson Hilbert space $\mathcal{B}_N$ is the *Bose condensate* $|B\rangle$, in which all N particles condense into the lowest-energy s.p. state, $b^\dagger_0|0\rangle$,

$$|\Phi_0\rangle \longrightarrow |B\rangle \equiv (N!)^{-\frac{1}{2}} (b^\dagger_0)^N |0\rangle \quad . \tag{29}$$

If we now specifically single out the condensate index 0 from the complete set of s.p. labels $\{\alpha\}$ in Sect. 2.1, as follows,

$$\alpha = \begin{cases} 0 & ; \text{ condensate state} \\ k \neq 0 & ; \text{ non-condensate states} \end{cases} , \tag{30}$$

we find that the set-index I becomes

$$I \rightarrow \left(m^I_{k_1}, m^I_{k_2}, \cdots\right) \quad ; \quad m^I_{k_i} = 0, 1, 2, \cdots \quad , \quad \forall i \quad , \tag{31}$$

in the notation of Sect. 2.1, and the multiconfigurational creation operators $C^\dagger_I$ can be written as follows,

$$C^\dagger_I \longrightarrow \left[\frac{(N-n)!}{N!}\right]^{\frac{1}{2}} \prod_i \left[\left(m^I_{k_i}!\right)^{-\frac{1}{2}} \left(b^\dagger_{k_i}\right)^{m^I_{k_i}} (b_0)^n\right] \quad , \tag{32}$$

where $\sum_i m^I_{k_i} = n$, and $n = 0, 1, 2, \cdots$. Alternatively, in the coordinate-space basis, the set-index I is as given by (24), and the generic multiconfigurational creation operators $C^\dagger_I$ become

$$C^\dagger_I \longrightarrow \left[\frac{(N-n)!}{N!}\right]^{\frac{1}{2}} \prod_{i=1}^{n} b^\dagger(\mathbf{r}_i)(b_0)^n \quad . \tag{33}$$

2.3 Many-Fermion System

We now consider a comparable system of N fermions, such as liquid ^{3}He, finite nuclei, nuclear matter, the electron gas, etc. In this case an obvious (but not unique) candidate for the model state $|\Phi_0\rangle$ in the fixed-N antisymmetric Hilbert space $\mathcal{F}_N$ is the *filled Fermi sea*, $|F\rangle$. This is an $(N \times N)$ Slater determinant of s.p. states formed from some complete s.p. basis $\{|\alpha_i\rangle\}$, usually from the N states of lowest energy. The single-fermion creation and destruction operators, $a^\dagger_\alpha$ and a_α, respectively, obey the usual anticommutation relations,

$$\{a_\alpha, a_\beta\} = 0 = \{a^\dagger_\alpha, a^\dagger_\beta\} \quad ; \quad \{a_\alpha, a^\dagger_\beta\} = \delta_{\alpha\beta}\mathbb{1} \quad , \tag{34}$$

and the s.p. states are obtained from the vacuum state $|0\rangle$ as,

$$|\alpha\rangle \equiv a^\dagger_\alpha|0\rangle \quad . \tag{35}$$

In terms of the s.p. creation operators we may write the antisymmetric state $|F\rangle$ in the second-quantised form,

$$|\Phi_0\rangle \longrightarrow |F\rangle = \prod_{i=1}^{N} a^\dagger_{\nu_i}|0\rangle \quad , \tag{36}$$

where we have adopted the standard notation,

$$\alpha_i = \begin{cases} \nu_i \ ; \ i = 1, 2, \cdots, N \ ; & \text{if } |\alpha_i\rangle \in |F\rangle \\ \rho_i \ ; \ i = 1, 2, \cdots \quad ; & \text{if } |\alpha_i\rangle \notin |F\rangle \end{cases} , \tag{37}$$

which is the fermionic analogue of the bosonic relation (30). Thus, we subdivide the s.p. states $|\alpha_i\rangle$ into two classes with repect to the model state $|F\rangle$, viz., those states $\{|\nu_i\rangle;\ i = 1, 2, \cdots, N\}$ from which $|F\rangle$ is constructed, and the remaining states $\{|\rho_i\rangle;\ i = 1, 2, \cdots\}$. For reasons in connection with creating new states from $|F\rangle$, the states $|\nu_i\rangle$ which are "occupied" in $|F\rangle$ are called hole states, and the states $|\rho_i\rangle$ which are "unoccupied" in $|F\rangle$ are called particle states. By analogy with the notation introduced in Sect. 2.1, the sets of particle-state indices $\{\rho_i\}$ and hole-state indices $\{\nu_i\}$ may be written as ordered sets, $\{\bar{\rho}_i;\ \bar{\rho}_1 < \bar{\rho}_2 < \cdots\}$ and $\{\bar{\nu}_i;\ \bar{\nu}_1 < \bar{\nu}_2 < \cdots < \bar{\nu}_N\}$ respectively.

In $\mathcal{F}_N$ we may now write the following equivalent completeness relations,

$$\begin{aligned} \mathbb{1} &= \frac{1}{N!} \sum_{\alpha_1, \cdots, \alpha_N} a^\dagger_{\alpha_1} \cdots a^\dagger_{\alpha_N} |0\rangle\langle 0| a_{\alpha_N} \cdots a_{\alpha_1} \\ &= \sum_{\bar{\alpha}_1 < \bar{\alpha}_2 < \cdots < \bar{\alpha}_N} a^\dagger_{\bar{\alpha}_1} \cdots a^\dagger_{\bar{\alpha}_N} |0\rangle\langle 0| a_{\bar{\alpha}_N} \cdots a_{\bar{\alpha}_1} \\ &= \sum_{m=0}^{N} \sum_{\bar{\rho}_1 < \bar{\rho}_2 < \cdots < \bar{\rho}_m} \sum_{\bar{\nu}_1 < \bar{\nu}_2 < \cdots < \bar{\nu}_m} a^\dagger_{\bar{\rho}_1} \cdots a^\dagger_{\bar{\rho}_m} a_{\bar{\nu}_1} \cdots a_{\bar{\nu}_m} |F\rangle\langle F| \\ &\qquad \times a^\dagger_{\bar{\nu}_m} \cdots a^\dagger_{\bar{\nu}_1} a_{\bar{\rho}_m} \cdots a_{\bar{\rho}_1} \\ &= \sum_{m=0}^{N} \frac{1}{(m!)^2} \sum_{\rho_1, \cdots, \rho_m} \sum_{\nu_1, \cdots, \nu_m} a^\dagger_{\rho_1} \cdots a^\dagger_{\rho_m} a_{\nu_1} \cdots a_{\nu_m} |F\rangle\langle F| \\ &\qquad \times a^\dagger_{\nu_m} \cdots a^\dagger_{\nu_1} a_{\rho_m} \cdots a_{\rho_1} \ . \end{aligned} \tag{38}$$

Hence, we see that with respect to the model state $|F\rangle$ the multiconfigurational creation operators $C^\dagger_I$ are products of particle–hole pair creation operators, $a^\dagger_{\rho_i} a_{\nu_i}$. More specifically the set-index I becomes

$$I \longrightarrow (n^I_{\bar{\nu}_1}, \cdots, n^I_{\bar{\nu}_N}; n^I_{\bar{\rho}_1}, n^I_{\bar{\rho}_2}, \cdots) \ ; \quad n^I_{\bar{\rho}_i}, n^I_{\bar{\nu}_j} = 0, 1 \ , \ \forall i, j \ , \tag{39}$$

and the multiconfigurational creation operators $C^\dagger_I$ become

$$C^\dagger_I \longrightarrow \prod_i \left(a^\dagger_{\bar{\rho}_i}\right)^{n^I_{\bar{\rho}_i}} \prod_{j=1}^{N} \left(a_{\bar{\nu}_j}\right)^{n^I_{\bar{\nu}_j}} \ ;$$

$$\sum_i n^I_{\bar{\rho}_i} = \sum_{j=1}^{N} n^I_{\bar{\nu}_j} = n \ ; \quad n = 0, 1, \cdots, N \ . \tag{40}$$

In the thermodynamic limit ($N \to \infty, \Omega \to \infty, \rho \equiv N/\Omega$ fixed), if the system remains homogeneous (and this is an assumption), the resulting translational invariance implies that the s.p. states are (best chosen as) plane waves,

$$|\alpha\rangle \longrightarrow |k\rangle \equiv |\mathbf{k}, i\rangle \quad , \tag{41}$$

where $i = 1, \cdots, \nu$ labels the internal quantum numbers (e.g., spin, isospin) and ν is the associated degeneracy. In this case,

$$\begin{aligned} |\rho\rangle &\longrightarrow |\mathbf{k}, i\rangle \quad ; \quad |\mathbf{k}| > k_F \\ |\nu\rangle &\longrightarrow |\mathbf{k}, i\rangle \quad ; \quad |\mathbf{k}| < k_F \quad , \end{aligned} \tag{42}$$

in terms of the *Fermi wavenumber*, k_F. Using the particle number operator, $\hat{N}$,

$$\hat{N} = \sum_{\mathbf{k},i} a^{\dagger}_{\mathbf{k}i} a_{\mathbf{k}i} \quad , \tag{43}$$

and (42), we easily prove that

$$\frac{N}{\Omega} \equiv \rho = \frac{\nu k_F^3}{6\pi^2} \quad . \tag{44}$$

It is worth noting that there also exist alternative choices to $|F\rangle$ for the model state $|\Phi_0\rangle$ for fermion systems. A common alternative is just the standard BCS state, $|S\rangle$, introduced by Bardeen, Cooper, and Schrieffer [88] in the context of superconductivity. The BCS state $|S\rangle$ is simply another Slater determinant analagous to $|F\rangle$, i.e., formed from quasiparticle states comparable to those in (35), but which are created by quasiparticle creation operators which are linear combinations of the previous particle and hole states. The corresponding Bogoliubov transformation is a canonical one, so that the quasiparticle creation and destruction operators obey the same fermion anticommutation relations (34).

2.4 Single Spin in an Irreducible Multiplet, J

It is important to realise that we can also handle, in the same broad general framework, systems of particles other than just bosons or fermions. Quantum-mechanical spins provide a good example. The simplest system of this type is just a single spin in an irreducible multiplet, J. In this case the Hilbert space $\mathcal{H}_J$ is now finite-dimensional and is spanned by the set of $(2J+1)$ states, $\{|J, M_J\rangle;\ M_J = -J, -J+1, \cdots, J\}$. The fundamental underlying algebra, SU(2), is defined by the usual commutation relations between the operators, $J^{\pm} \equiv J^x \pm \mathrm{i}J^y$, and J^z,

$$[J^z, J^{\pm}] = \pm J^{\pm} \quad ; \quad [J^+, J^-] = 2J^z \quad . \tag{45}$$

We note that models of this category arise in various guises. One example is the well-known model many-body Hamiltonian of Lipkin, Meshkov, and Glick [89] (the LMG model), which was originally invented to simulate the collective monopole vibrations in spherical atomic nuclei. The original model comprises N identical fermions distributed between two energy levels, each of

which is N-fold degenerate, and interacting via a simple pairing interaction. The interest in the LMG model and its variants, centres on the fact that they exhibit a phase or shape transition at some critical value of the coupling strength. In nuclear physics, the transition from spherical to deformed shape which occurs in the regime of the rare-earth nuclei is believed to be due to a collective quadrupole excitation of this type, and the LMG model was first introduced as its simpler monopole analogue.

A second reason for studying such anharmonic spin models is that they provide a finite-dimensional Hilbert space analogue of the corresponding anharmonic oscillator models which have infinite-dimensional Hilbert spaces, and whose mathematical properties can thereby be obscured. Thus, it is easy to verify that if we define a new set of operators,

$$\mathcal{J}_0 \equiv J^z + J \;\; ; \quad \mathcal{J}_\pm \equiv (2J)^{-\frac{1}{2}} J^\pm \;\; , \tag{46}$$

the commutation relations for large values of J between the operators $\mathcal{J}_+, \mathcal{J}_-$, and $\mathcal{J}_0$ are the same as those between bosonic operators $b^\dagger, b$, and $b^\dagger b$, respectively, except for terms of order $O(\langle b^\dagger b \rangle / J)$. Hence, for example, in the limit that $J \rightarrow \infty$, the anharmonic spin Hamiltonian, H_J, defined as

$$H_J = \frac{1}{2} + J + J^z + \frac{\lambda}{16J^2}(J^+ + J^-)^4 \;\; , \tag{47}$$

should, making use of (28), smoothly approach the anharmonic oscillator Hamiltonian,

$$\begin{aligned} H &= b^\dagger b + \frac{1}{2} + \frac{\lambda}{4}(b^\dagger + b)^4 \\ &= \frac{1}{2}\hat{p}^2 + \frac{1}{2}\hat{x}^2 + \lambda \hat{x}^4 \;\; , \end{aligned} \tag{48}$$

in the well-defined sense that any energy eigenvalue $E_n^{(J)}$ with fixed value of n should converge to the corresponding anharmonic oscillator energy eigenvalue E_n. Aalto *et al.* [90] have performed an ECCM analysis of both Hamiltonians (47) and (48).

We now consider the various possible choices of model state $|\Phi_0\rangle$ from the $(2J+1)$ independent states spanning $\mathcal{H}_J$.

Lowest State, $|\Phi_0\rangle \rightarrow |J, -J\rangle$: From the basic commutation relations (45) one may easily prove that

$$J^+|J, -J+m\rangle = [(m+1)(2J-m)]^{\frac{1}{2}}|J, -J+m+1\rangle \;\; ; \quad m = 0, 1, \cdots, 2J \;\; . \tag{49}$$

Equation (49) gives immediately that, in relation to the model state $|\Phi_0\rangle \rightarrow |J, -J\rangle$, the generic operators $C_I^\dagger$ take the following form,

$$C_I^\dagger \longrightarrow \left[\frac{(2J-m)!}{(2J)!m!}\right]^{\frac{1}{2}} (J^+)^m \;\; , \tag{50}$$

where the label I takes the values,

$$I \longrightarrow m \;\; ; \quad m = 0, 1, \cdots, 2J \;\; . \tag{51}$$

Highest State, $|\Phi_0\rangle \rightarrow |J, J\rangle$: In this case the role of the basic creation operator is now taken by $J^- \equiv (J^+)^\dagger$, and otherwise everything now goes exactly as in the previous case, but with $J^+ \rightarrow J^-$.

Intermediate State, $|\Phi_0\rangle \rightarrow |J, M\rangle$, with $M \neq \pm J$: In this case, although we can clearly construct all remaining states in $\mathcal{H}_J$ by letting either J^+ or J^- act on $|\Phi_0\rangle$ a sufficient number of times, we cannot now construct two Abelian subalgebras from their necessarily simultaneous use to span $\mathcal{H}_J$, since $[J^+, J^-] \neq 0$. Nevertheless, it is important to realise that this does *not* imply that such intermediate states cannot be chosen as model states. Rather, we need to redefine our creation operators, $C_I^\dagger$, accordingly. As a specific example of how this may be done more generally, let us consider the case $J = 1$, and attempt to use $|\Phi_0\rangle = |1, 0\rangle$ as a model state. If we now use the column vector notation for the angular momentum eigenstates,

$$|1,1\rangle \equiv \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \;\; ; \quad |1,0\rangle \equiv \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \;\; ; \quad |1,-1\rangle \equiv \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \;\; , \tag{52}$$

we may define two new operators as follows,

$$U^+ \equiv \begin{pmatrix} 0\,1\,0 \\ 0\,0\,0 \\ 0\,0\,0 \end{pmatrix} \;\; ; \quad D^+ \equiv \begin{pmatrix} 0\,0\,0 \\ 0\,0\,0 \\ 0\,1\,0 \end{pmatrix} \;\; , \tag{53}$$

whose mode of action on the model state is

$$U^+|1,0\rangle = |1,1\rangle \;\; ; \quad D^+|1,0\rangle = |1,-1\rangle \;\; . \tag{54}$$

However, whereas their mode of action on $|1,0\rangle$ is thus identical to those of J^+ and J^-, respectively, the new operators now *do* commute, $[U^+, D^+] = 0$. Thus, we may use $|\Phi_0\rangle = |1,0\rangle$ as a model state in terms of the set of mutually commuting creation operators,

$$\{C_I^\dagger\} \longrightarrow \{\mathbb{1}, U^+, D^+\} \;\; . \tag{55}$$

2.5 Quantum Spin Lattices, e.g., Antiferromagnets

In the case of several similar spins, we may simply take the tensor products of the operators introduced in Sect. 2.4 for our multiconfigurational creation operators, $C_I^\dagger$. Some examples are given below.

Spin-Half Particles on a Bipartite Lattice: A bipartite lattice is one on which there are two completely equivalent sublattices A and B such that all nearest neighbours of sites on sublattice A lie on sublattice B, and vice versa (e.g., a square lattice in two spatial dimensions, and a simple cubic lattice in three dimensions). On such lattices the *classical* ground state of many Hamiltonians is often the ideal Néel antiferromagnetic state in which all spins on sublattice A align along one direction (say, the negative z-direction) and all spins on sublattice B align along the opposite (i.e., positive z) direction. A simple example is provided by the Heisenberg Hamiltonian,

$$H = J \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j \quad , \tag{56}$$

where the sum on $\langle i,j \rangle$ denotes a sum over all nearest-neighbour pairs, and $\mathbf{s}_i$ is the spin on site i, in the case where the exchange constant, $J > 0$. In the quantum-mechanical case the interest in such models is often concerned with whether the quantum fluctuations (or quantum correlations) will or will not completely destroy the perfect Néel long-range order of the classical antiferromagnet.

In such cases, an appropriate choice of model state $|\Phi_0\rangle$ for spin-half particles is the Néel state $|N\rangle$, which is simply the tensor product,

$$|N\rangle \equiv \bigotimes_{i \in A} |\downarrow\rangle_i \bigotimes_{j \in B} |\uparrow\rangle_j \quad ; \tag{57}$$

of all "down" states, $|\downarrow\rangle \equiv |\frac{1}{2}, -\frac{1}{2}\rangle$, on sites i of sublattice A with all "up" states, $|\uparrow\rangle \equiv |\frac{1}{2}, \frac{1}{2}\rangle$, on sites j of sublattice B. The configuration label (or set-index) I may now be taken as the set of sites,

$$I \longrightarrow (i_1, \cdots, i_m; j_1, \cdots, j_n) \quad ; \quad m, n = 0, 1, \cdots, \tag{58}$$

on which the spins are reversed with respect to the Néel state $|N\rangle$. The mutually commuting set of multiconfigurational creation operators with respect to $|N\rangle$ may thus be taken as,

$$C_I^\dagger \longrightarrow s_{i_1}^+ \cdots s_{i_m}^+ s_{j_1}^- \cdots s_{j_n}^- \quad . \tag{59}$$

We note again that the choice $|N\rangle$ for the model state $|\Phi_0\rangle$ is not unique. In cases where the ground state of the quantum Hamiltonian is expected to be, for example, dimerised or trimerised, one may construct model states $|\Phi_0\rangle$ built from products of suitable (noninteracting) dimer or trimer states. The interested reader is referred to the literature [91,92] for further details of such states, which are particularly pertinent for a valence-bond description of quantum spin lattices.

Spin-One Particles on a Bipartite Lattice: In the case of spin-one particles, the Néel state (57) is still a possible choice of model state $|\Phi_0\rangle$, where $|\downarrow\rangle \equiv |1,-1\rangle$. However, the multiconfigurational creation operators (59) now include the possibility that a single-spin raising operator on a site $i \in A$ or a single-spin lowering operator on a site $j \in B$ may appear either once or twice on the same site, in order to include all three possible spin projections on each site. Similarly, the configuration label (or set-index) I given by (58) must be extended to allow any given site index to appear up to two times in the set.

By extension of the discussion in Sect. 2.4 we may also consider an alternative planar model state $|\Phi_0\rangle \rightarrow |P\rangle$, where $|P\rangle$ is the state in which the spin on every site is in the state $|0\rangle \equiv |1,0\rangle$ with zero projection along the quantisation z-axis. In this case a suitable set of mutually commuting multiconfigurational creation operators C_I^+ is given by the tensor product of operators U_k^+ and D_k^+ given by (53) over a given subset of sites $\{k\}$. The set-index label I now comprises the subset $\{k\}$, together with a two-valued flag at each member of the subset which indicates whether the operator U_k^+ or D_k^+ acts at each site. Once again, many other more complicated model states, $|\Phi_0\rangle$, can also be devised.

3 Basic Structure of the CCM

After the discussion in Sect. 2 we are now in a position to explain the basic ingredients and the formal structure of the CCM at a very general level, without reference to specific many-body systems.

3.1 Exponential Structure of Many-Body Wave Functions

Exponential structures arise frequently in physics, and often for similar underlying fundamental reasons. For example, in statistical mechanics, the Ursell–Mayer cluster expansion for the thermodynamic partition function (from which all thermodynamic properties of a bulk system derive) has an underlying *exponential form.* This arises naturally from the basic compounding, *with the correct statistical multiplicities,* of all contributions to the partition function from the *independent* "atomic clusters" of a given size.

We shall now show that a comparable exponential form also underpins the natural parametrisation (i.e., the "correct" form) for a quantum-mechanical many-body wave function. Historically, this was probably first realised by Goldstone [93] and Hubbard [94] in the context of time-independent perturbation theory (TIPT), where it also takes the related form of the *Goldstone linked cluster theorem.* Similar ideas apply equally well to the well-known representation of eigenstates in quantum field theory provided by the Gell-Mann and Low theorem [95], although this seems not to have been properly or fully realised at that time.

For pedagogical purposes let us consider a "closed-shell" N-fermion system, i.e., one which is reasonably approximated in zeroth order by a s.p. shell-model Slater determinant $|\Phi_0\rangle = |F\rangle$ of the sort discussed in Sect. 2.3, and which can be expressed in the second-quantised form (36). The choice of s.p. states from which to construct this filled Fermi sea is, at this stage, completely arbitrary. For example, one may choose harmonic oscillator states, self-consistent Hartree-Fock states, or any other basis. In this context an important theorem of Thouless [96] asserts that the most general determinantal wave function $|\Phi_0'\rangle$ not orthogonal to a given Slater determinant $|\Phi_0\rangle$ has the form

$$|\Phi_0'\rangle = \mathrm{e}^{S_1}|\Phi_0\rangle \quad , \tag{60}$$

for some suitable choice of the one-body operator S_1 which acts on $|\Phi_0\rangle$ to produce a one-particle/one-hole ($1p$–$1h$) excitation. Thus, in the notation of Sect. 2.3, we may write

$$S_1 = \sum_{\rho} \sum_{\nu} \langle \rho | S_1 | \nu \rangle \, a_\rho^\dagger a_\nu \quad . \tag{61}$$

The effect of allowing single particles to be independently elevated above the Fermi sea (which, as we shall see below, is what is encapsulated in the exponential operation in the relation (60)) is, thus, equivalent to changing the s.p. orbitals that comprise the Slater determinant.

Consider now the problem of obtaining the exact ground-state wave function, $|\Psi_0\rangle$, and energy, E_0, of the interacting system of fermions described by the many-body Hamiltonian, H,

$$H|\Psi_0\rangle = E_0|\Psi_0\rangle \quad . \tag{62}$$

The *dynamical correlations* induced by the interactions contained in H will modify the model state $|\Phi_0\rangle$. We *assume* from the outset that $|\Psi_0\rangle$ has a nonzero overlap with $|\Phi_0\rangle$, and we shall henceforth work in the *intermediate normalisation* scheme wherein

$$\langle \Phi_0 | \Psi_0 \rangle = \langle \Phi_0 | \Phi_0 \rangle \equiv 1 \quad . \tag{63}$$

Now, the simplest correlation, other than a single $1p$–$1h$ excitation discussed above in the context of the Thouless theorem, is a single two-particle/two-hole ($2p$–$2h$) excitation of the form $S_2|\Phi_0\rangle$, where

$$S_2 = \frac{1}{(2!)^2} \sum_{\rho_1,\rho_2} \sum_{\nu_1,\nu_2} \langle \rho_1 \rho_2 | S_2 | \nu_1 \nu_2 \rangle_A \, a_{\rho_1}^\dagger a_{\rho_2}^\dagger a_{\nu_2} a_{\nu_1} \quad , \tag{64}$$

and the suffix A on the matrix element $\langle \rho_1 \rho_2 | S_2 | \nu_1 \nu_2 \rangle_A$ denotes an antisymmetrised element (in the notation where $|\nu_1 \nu_2\rangle_A \equiv |\nu_1 \nu_2\rangle - |\nu_2 \nu_1\rangle$).

Now, it may also happen that two *independent* pairs of particles also excite out of $|\Phi_0\rangle$ due to dynamic correlations. Clearly, this process is described by applying the operator S_2 twice, but with the proviso that we must include

a statistical weighting factor (or multiplicity) of $\frac{1}{2}$ to avoid double counting. The resulting contribution to the correlated wave function is $\frac{1}{2}S_2^2|\Phi_0\rangle$. This process of independent excitation of pairs out of the Fermi sea may be continued to obtain a contribution $(1/m!)S_2^m|\Phi_0\rangle$ from the excitation of m independent $2p$–$2h$ pairs. Hence, the total amplitude for the excitation of an arbitrary number (including zero) of independent $2p$–$2h$ excitations is

$$\sum_{m=0}^{\infty} \frac{1}{m!} S_2^m |\Phi_0\rangle = e^{S_2}|\Phi_0\rangle \quad , \tag{65}$$

and we note, very importantly, that the exponential structure arises very simply and fundamentally from a simple *counting of independent events.* This exponential structure lies at the very heart of the CCM.

Next we consider processes involving the simultaneous excitation of three fermions from the Fermi sea. These may be described by a contribution $S_3|\Phi_0\rangle$ to the exact wave function. Similarly to the case of pairs, there will be a contribution $(1/n!)S_3^n|\Phi_0\rangle$ from the simultaneous excitation of n independent triplets. If we then take into account the possibility of the independent simultaneous excitation of m pairs and n triplets from the Fermi sea, the corresponding contribution to $|\Psi_0\rangle$ is $(1/m!n!)S_2^m S_3^n|\Phi_0\rangle$. It is important that we do not need to worry about the order of the operators S_2 and S_3 in this product because they describe independent processes and hence commute, as one can also verify from their specific second-quantised forms. Summing over all possible values of m and n then leads to the contribution $\exp(S_2+S_3)|\Phi_0\rangle$ to $|\Psi_0\rangle$. Proceeding in this way with the independent excitation of clusters of up to N particles we arrive at the CCM representation,

$$|\Psi_0\rangle = e^S|\Phi_0\rangle \quad ; \quad S = \sum_{m=1}^{N} S_m \quad , \tag{66a}$$

$$S_m = \frac{1}{(m!)^2} \sum_{\rho_1 \cdots \rho_m} \sum_{\nu_1 \cdots \nu_m} \langle \rho_1 \cdots \rho_m | S_m | \nu_1 \cdots \nu_m \rangle_A \, a_{\rho_1}^\dagger \cdots a_{\rho_m}^\dagger a_{\nu_m} \cdots a_{\nu_1} \quad . \tag{66b}$$

3.2 The Configuration-Interaction Method (CIM)

For the same N-fermion case considered above, a more naive and (seemingly) simpler parametrisation of $|\Psi_0\rangle$ is that used in the CIM,

$$|\Psi_0\rangle = (1+F)|\Phi_0\rangle \quad ; \quad F = \sum_{m=1}^{\infty} F_m \quad , \tag{67}$$

$$F_m = \frac{1}{(m!)^2} \sum_{\rho_1 \cdots \rho_m} \sum_{\nu_1 \cdots \nu_m} \langle \rho_1 \cdots \rho_m | F_m | \nu_1 \cdots \nu_m \rangle_A \, a_{\rho_1}^\dagger \cdots a_{\rho_m}^\dagger a_{\nu_m} \cdots a_{\nu_1} \quad . \tag{68}$$

Again, F_n excites n particles and n holes from the filled Fermi sea $|\Phi_0\rangle \equiv |F\rangle$, but in this case F_n generally contains *unlinked* (or *disconnected*) pieces which arise from the products of noninteracting subclusters. A comparison of the first few terms in the expansions (65) and (67) yields, for example,

$$F_1 = S_1 \ ; \ F_2 = S_2 + \frac{1}{2}S_1^2 \ ; \ F_3 = S_3 + S_2S_1 + \frac{1}{6}S_1^3 \ ;$$

$$F_4 = S_4 + S_3S_1 + \frac{1}{2}S_2^2 + \frac{1}{2}S_2S_1^2 + \frac{1}{24}S_1^4 \ ; \ \text{etc.} \ , \tag{69}$$

with corresponding inverses,

$$S_1 = F_1 \ ; \ S_2 = F_2 - \frac{1}{2}F_1^2 \ ; \ S_3 = F_3 - F_2F_1 + \frac{1}{3}F_1^3 \ ;$$

$$S_4 = F_4 - F_3F_1 - \frac{1}{2}F_2^2 + F_2F_1^2 - \frac{1}{4}F_1^4 \ ; \ \text{etc.} \tag{70}$$

More generally, by putting $F_n \to \lambda^n F_n$ and $S_n \to \lambda^n S_n$, and by equating powers of λ in the two expansions (65) and (67), we find

$$F_n = \sum_{l=1}^{n} \sum_{m_1} \cdots \sum_{m_l} \frac{1}{m_1! \cdots m_l!} S_1^{m_1} \cdots S_l^{m_l} \ ; \ \sum_{i=1}^{l} i m_i = n \ , \tag{71}$$

where the restriction on the sums over $m_1, \cdots, m_l$ is such that one sums over all sets of non-negative integers $\{m_i \geq 0\}$ such that $\sum_{i=1}^{l} i m_i = n$. Since the number of ways of arranging k identical objects into $l \leq k$ classes is $k!/(m_1! \cdots m_l!)$, where m_i objects are in class i, and $\sum_i m_i = k$, we easily derive the alternative relation,

$$F_n = \sum_{k=1}^{n} \frac{1}{k!} \sum_{n_1} \cdots \sum_{n_k} S_{n_1} \cdots S_{n_k} \ ; \ \sum_{i=1}^{k} n_i = n \ , \tag{72}$$

where the restriction on the sums over $n_1, \cdots, n_k$ is such that one sums over all sets of positive integers $\{n_i > 0\}$ such that $\sum_{i=1}^{k} n_i = n$. The corresponding inverse relations are also readily found by using $S = \ln(1+F)$. The analogues to (71) and (72) are thus,

$$S_n = \sum_{l=1}^{n} \sum_{m_1} \cdots \sum_{m_l} (-1)^{k-1} \frac{(k-1)!}{m_1! \cdots m_l!} F_1^{m_1} \cdots F_l^{m_l} \ ;$$

$$\sum_{i=1}^{l} i m_i = n \ , \ \sum_{i=1}^{l} m_i = k \ , \tag{73}$$

and

$$S_n = \sum_{k=1}^{n} \frac{(-1)^{k-1}}{k} \sum_{n_1} \cdots \sum_{n_k} F_{n_1} \cdots F_{n_k} \ ; \ \sum_{i=1}^{k} n_i = n \ . \tag{74}$$

We note that both the CCM and CIM parametrisations are exact and equivalent so long as no approximations (i.e., no truncations over the complete set of multiparticle configurations) are made. However, once we approximate the equivalence is lost. We note that whereas, by construction, all of the matrix elements of the operators S_n are linked, the matrix elements of the operators F_n mix linked with unlinked terms (e.g., F_4 mixes the linked $4p$–$4h$ term S_4 with unlinked terms like $\frac{1}{2}S_2^2$ which corresponds to two unlinked $2p$–$2h$ pairs). Although the unlinked terms ultimately cancel out in any exact theory, this is not true in the CIM when truncations are made. By contrast, the cancellation of unlinked terms is automatic in the CCM, whatever approximations are made to the *cluster correlation operator, S*.

In practice the CIM just amounts to truncating the number of possible many-body configurations excited out of $|\Phi_0\rangle$ to some number $\mathcal{N}$, and then diagonalising the Hamiltonian as an $\mathcal{N} \times \mathcal{N}$ matrix in this truncated multiconfigurational basis. The hope is then that as $\mathcal{N}$ is increased one obtains convergence. As we have explained in Sect. 1.2 the interlacing theorem [51] guarantees this (uniform) convergence in principle. However, for large numbers of particles ($N \rightarrow \infty$) the matrix becomes *ill-conditioned*, and the CIM becomes fatally flawed due to the inexact cancellation of all unlinked terms. What happens is that for such extensive variables as the energy, E_0, the linked terms lead properly to contributions which scale (properly) linearly in the particle number, N. By contrast terms containing m unlinked pieces scale as N^m, and the (in principle) exact cancellation of contributions from terms with $m \geq 2$ rapidly becomes numerically unstable with increasing N, unless the exact cancellation is incorporated from the outset, as in the CCM. These ideas are expressed more quantitatively later, but are perhaps first best related to the so-called *"size-extensivity problem"* of the CIM [52], already alluded to in Sect. 1.2.

3.3 Size-Extensivity

Consider a system of N particles, whose ground-state wave function $|\Psi_0\rangle$ is parametrised by operators $\{F_n\}$ in the CIM and $\{S_n\}$ in the CCM. Suppose we now separate the system spatially into two subsystems A and B, with N_A particles in A and N_B particles in B. We assume that as the spatial separation increases, $r_{AB} \rightarrow \infty$, the two subsystems cease to interact with each other, so that the Hamiltonian becomes,

$$H \underset{r_{AB}\rightarrow\infty}{\longrightarrow} H^A + H^B \quad , \quad \text{with } [H^A, H^B] = 0 \quad . \tag{75}$$

Hence, in this limit, the energy becomes additively separable,

$$E_0 \underset{r_{AB}\rightarrow\infty}{\longrightarrow} E_0^A + E_0^B \quad , \tag{76}$$

while the wave function becomes multiplicatively separable,

$$|\Psi_0\rangle \underset{r_{AB}\to\infty}{\longrightarrow} |\Psi_0^A\rangle \otimes |\Psi_0^B\rangle \ . \tag{77}$$

A system is said to be *size-extensive* when it obeys the above separability properties (76) and (77).

We can now see immediately that the exponential parametrisation of the wave function given by (65) in the CCM guarantees the separability relation (77), no matter how the cluster correlation operator S is truncated. For example, a common truncation scheme in both the CCM and CIM is the so-called SUBn approximation scheme, wherein the m-body partitions of the operator S in (65) or of the operator F in (67) are truncated so that one retains only the components with $m \leq n$, and sets the higher partitions with $m > n$ to zero. Thus, in the CCM SUBn scheme we have,

$$\begin{aligned} |\Psi_0\rangle &\approx \exp(S_1 + \cdots + S_n)|\Phi_0\rangle \\ &\underset{r_{AB}\to\infty}{\longrightarrow} \exp(S_1^{(A)} + S_1^{(B)} + \cdots + S_n^{(A)} + S_n^{(B)})|\Phi_0\rangle \\ &\equiv \exp(S_1^{(A)} + \cdots + S_n^{(A)})|\Phi_0^A\rangle \otimes \exp(S_1^{(B)} + \cdots + S_n^{(B)})|\Phi_0^B\rangle \ . \end{aligned} \tag{78}$$

By contrast, in the CIM SUBn scheme, we have

$$\begin{aligned} |\Psi_0\rangle &\approx (1 + F_1 + \cdots + F_n)|\Phi_0\rangle \\ &\underset{r_{AB}\to\infty}{\not\longrightarrow} (1 + F_1^{(A)} + \cdots + F_n^{(A)})|\Phi_0^A\rangle \otimes (1 + F_1^{(B)} + \cdots + F_n^{(B)})|\Phi_0^B\rangle \ . \end{aligned} \tag{79}$$

In this case the separability of the wave function is *not* preserved since the separated parametrisation in the second line of (79) would require excitations of up to $2n$ particle/hole pairs which are not contained in a SUBn scheme.

3.4 General Cumulant (or Connected Moment) Expansions

The exponential parametrisation (65) of the CCM is intimately connected with similar exponential structures underpinning general *cumulant expansions*. Thus, in probability theory one considers the moment generating function, $\phi(\lambda)$, defined as

$$\phi(\lambda) \equiv \langle \exp(\lambda x) \rangle = \sum_{n=0}^{\infty} \frac{\lambda^n}{n!} \mu_n \ ,$$

$$\mu_n \equiv \langle x^n \rangle \ , \ n = 1, 2, \cdots \ ; \ \mu_0 \equiv 1 \ . \tag{80}$$

The function $\phi(\lambda)$ thereby generates all of the positive integral *moments*, $\mu_n \equiv \langle x^n \rangle$, of an *arbitrary random variable*, x, according to some *probability distribution* implied by the angular brackets. This average, $\langle \cdots \rangle$, can either be a "classical", e.g., thermal, average or a quantal expectation value, e.g., $x \to H$, and $\langle \cdots \rangle \to \langle \Phi_0 | \cdots | \Phi_0 \rangle$ gives the energy moments $\langle \Phi_0 | H^n | \Phi_0 \rangle$ in the state $|\Phi_0\rangle$. In practice, the state $|\Phi_0\rangle$ is an arbitrary trial state, which

is required only to have a nonzero overlap with the exact ground state $|\Psi_0\rangle$, and which (thus usually) shares its principle symmetries.

In principle, the moments contain all of the information about the probability distribution. For example, the energy moments, $\mu_n \equiv \langle\Phi_0|H^n|\Phi_0\rangle, n \geq 1$, contain all of the dynamical information about the system, and one might expect to be able to construct all other quantities pertaining to the system from them. However, there are two important points to note about the moments, $\mu_n \equiv \langle x^n\rangle$, namely:
– they contain essentially redundant information, and
– the information is encapsulated in a very inefficient and obscure way.
These problems lead in practice to the fact that many naive inversion (or inversion-type) methods based on moments are badly ill-conditioned. This manifests itself in the problem that as one attempts to gain more information by using more moments, numerical rounding errors rapidly grow and frustrate these attempts, no matter what level of numerical precision is adopted.

A standard way to improve this situation is to use, instead of the moments themselves, the so-called *cumulants* (or, synonymously, the *connected moments* or *semi-invariants*) defined by [97],

$$\ln\langle\exp(\lambda x)\rangle \equiv \sum_{n=1}^{\infty} \frac{\lambda^n}{n!}\nu_n \quad , \tag{81}$$

where $\nu_n \equiv \langle x^n\rangle_c$ are the connected moments or cumulants. Such connected moments have many simple properties. Especially important are:
– that they scale linearly with the size of the system (i.e., $\nu_n \propto N$, $\forall n$, as $N \to \infty$) or, equivalently,
– that the combined cumulants for two *independent* subsystems are the corresponding sums of the two separate cumulants.
This latter relation is trivial to prove. Thus, for two random variables x and y we have, in an obvious notation,

$$\ln\langle\exp[\lambda(x+y)]\rangle \equiv \sum_{n=1}^{\infty} \frac{\lambda^n}{n!}\nu_n^{(x+y)} \quad . \tag{82}$$

However, if x and y are independent we have

$$\langle\exp[\lambda(x+y)]\rangle = \langle\exp(\lambda x)\rangle\langle\exp(\lambda y)\rangle \quad , \tag{83}$$

and hence,

$$\nu_n^{(x+y)} = \nu_n^{(x)} + \nu_n^{(y)}, \quad \forall n \quad . \tag{84}$$

Similarly, for k *independent* variables $\{x_i;\ i = 1, \cdots, k\}$, we have

$$\nu_n^{(x_1+\cdots+x_k)} = \sum_{i=1}^{k} \nu_n^{(x_i)}, \quad \forall n \quad , \tag{85}$$

and all cross-terms vanish.

By direct comparison of the coefficients of equal powers of λ in both series (80) and (81), one readily checks that the lowest n cumulants determine all moments up to order n, and vice versa. The explicit relationships between the first few are as follows,

$$\mu_1 = \nu_1 \;\; ; \;\; \mu_2 = \nu_2 + \nu_1^2 \;\; ; \;\; \mu_3 = \nu_3 + 3\nu_2\nu_1 + \nu_1^3 \;\; ;$$
$$\mu_4 = \nu_4 + 4\nu_3\nu_1 + 3\nu_2^2 + 6\nu_2\nu_1^2 + \nu_1^4 \;\; ; \;\; \text{etc.} \;\; , \tag{86}$$

with corresponding inverses,

$$\nu_1 = \mu_1 \;\; ; \;\; \nu_2 = \mu_2 - \mu_1^2 \;\; ; \;\; \nu_3 = \mu_3 - 3\mu_2\mu_1 + 2\mu_1^3 \;\; ;$$
$$\nu_4 = \mu_4 - 4\mu_3\mu_1 - 3\mu_2^2 + 12\mu_2\mu_1^2 - 6\mu_1^4 \;\; ; \;\; \text{etc.} \;\; , \tag{87a}$$

or, equivalently,

$$\langle x\rangle_c = \langle x\rangle \;\; ; \;\; \langle x^2\rangle_c = \langle x^2\rangle - \langle x\rangle^2 \;\; ; \;\; \langle x^3\rangle_c = \langle x^3\rangle - 3\langle x^2\rangle\langle x\rangle + 2\langle x\rangle^3 \;\; ;$$
$$\langle x^4\rangle_c = \langle x^4\rangle - 4\langle x^3\rangle\langle x\rangle - 3\langle x^2\rangle^2 + 12\langle x^2\rangle\langle x\rangle^2 - 6\langle x\rangle^4 \;\; ; \;\; \text{etc.} \tag{87b}$$

Clearly, by comparison of (67) and (65) with (80) and (81), respectively, we see that there is a complete analogy with the CIM and CCM representations,

$$\mu_n \longleftrightarrow n!F_n \;\; ; \quad \nu_n \longleftrightarrow n!S_n \;\; . \tag{88}$$

By comparison with the results derived in Sect. 3.2, we may easily prove the explicit general relationships,

$$\mu_n = \sum_{k=1}^{n} \frac{1}{k!} \sum_{n_1} \cdots \sum_{n_k} \frac{n!}{n_1! \cdots n_k!} \nu_{n_1} \cdots \nu_{n_k} \;\; ; \quad \sum_{i=1}^{k} n_i = n$$
$$= n! \sum_{l=1}^{n} \sum_{m_1} \cdots \sum_{m_l} \prod_{i=1}^{l} \left[\frac{1}{m_i!} \left(\frac{\nu_i}{i!} \right)^{m_i} \right] \;\; ; \quad \sum_{i=1}^{l} i m_i = n \;\; , \tag{89}$$

and the corresponding inverses,

$$\nu_n = \sum_{k=1}^{n} \frac{(-1)^{k-1}}{k} \sum_{n_1} \cdots \sum_{n_k} \frac{n!}{n_1! \cdots n_k!} \mu_{n_1} \cdots \mu_{n_k} \;\; ; \quad \sum_{i=1}^{k} n_i = n$$
$$= n! \sum_{l=1}^{n} \sum_{m_1} \cdots \sum_{m_l} (-1)^{k-1}(k-1)! \prod_{i=1}^{l} \left[\frac{1}{m_i!} \left(\frac{\mu_i}{i!} \right)^{m_i} \right] \;\; ;$$
$$\sum_{i=1}^{l} i m_i = n \;\; , \quad \sum_{i=1}^{l} m_i = k \;\; , \tag{90}$$

and where the summation variables are integers such that $n_i > 0$ and $m_i \geq 0$.

It is also not difficult to prove the recurrence relation,

$$\mu_{n+1} = \sum_{m=0}^{n} \frac{n!}{m!(n-m)!} \mu_{n-m} \nu_{m+1} \quad ; \quad \mu_0 = 1 \quad , \tag{91}$$

and its counterpart relating the CIM to the CCM correlation operators,

$$F_n = \sum_{m=1}^{n} \frac{m}{n} F_{n-m} S_m \quad ; \quad F_0 = \mathbb{1} \quad . \tag{92}$$

Relations (91) and (92) can be useful for numerical computations.

Such cumulants (or, equivalently the linked cluster correlation operators $\{S_n\}$ and their analogues) appear in many places in quantum many-body theory. They lie, for example, at the heart of the Goldstone linked cluster theorem [93], which we prove very easily below from within the CCM framework. More generally, they always lie at the heart of any classical or quantum-mechanical (diagrammatic or other) expansion which deals with the (physical) linked or connected quantities. In this very real sense the CCM parametrisation of ground-state wave functions is the quantal (i.e., operatorial) analogue of the classical (i.e., c-number) Ursell–Mayer [98] expansion for the partition function of classical statistical mechanics.

3.5 Formal Elements of the Single-Reference Normal CCM

By making use of the ideas and powerful notation introduced in Sect. 2, we are now in a position to present the key ingredients of the normal CCM (NCCM) formalism at a very general level. We restrict ourselves in the first instance to the single-reference (or "closed shell") version of the method, and to its zero-temperature formulation in terms of pure states.

Ket Ground State: In terms of a general model state, $|\Phi_0\rangle$, which plays the role of a cyclic vector with respect to a complete set of mutually commuting multiconfigurational creation operators, $C_I^\dagger$, as discussed in detail in Sect. 2, we now easily generalise the CCM parametrisation (66a,b) of a many-fermion ground state to the generic case. Thus, the exact many-body ket ground-state wave function (or, more generally, any exact ket eigenstate $|\Psi_0\rangle$ not orthogonal to $|\Phi_0\rangle$) is written as

$$|\Psi_0\rangle = \mathrm{e}^S |\Phi_0\rangle \quad ; \quad S = \sum_I{}' s_I C_I^\dagger \quad , \tag{93}$$

where the prime on the sum excludes the term $I = 0$ (corresponding to the case $C_0^\dagger = \mathbb{1}$), in order to preserve the intermediate normalisation scheme of (63). The operator S is a linked-cluster operator, by construction, and the quantities $\{s_I\}$ are a complete set of c-number cluster amplitudes. In general, $|\Phi_0\rangle$ will contain the important symmetries of the phase of the system under consideration, including the correct particle statistics, as appropriate.

Ground-State Energy: Insertion of the CCM parametrisation (93) into the ground-state Schrödinger equation (62) leads to the typical CCM form,

$$(\mathrm{e}^{-S}H\mathrm{e}^{S} - E_0)|\Phi_0\rangle = 0 \ , \tag{94}$$

in which the Hamiltonian appears in the similarity-transformed form which is a hallmark of the method. By taking the inner products of (94) with $|\Phi_0\rangle$ itself and with the remainder of the complete set of states $\{C_I^\dagger|\Phi_0\rangle; \ I \neq 0\}$ we are thus led respectively to an equation for $E_0 = E_0(s_I)$,

$$E_0 = \langle\Phi_0|\mathrm{e}^{-S}H\mathrm{e}^{S}|\Phi_0\rangle = \langle\Phi_0|H\mathrm{e}^{S}|\Phi_0\rangle \ , \tag{95}$$

and a set of formally exact, microscopic, coupled nonlinear equations for the cluster correlation coefficients $\{s_I\}$,

$$\langle\Phi_0|C_I\mathrm{e}^{-S}H\mathrm{e}^{S}|\Phi_0\rangle = 0 \ ; \ \forall I \neq 0 \ , \tag{96}$$

in which there appear no macroscopic quantities like the energy E_0. Just as for the specific case of a many-fermion system discussed in Sect. 3.3, so in the general case the energy eigenvalue, E_0, is guaranteed to be an extensive quantity, no matter how the formally exact sum over configurations $\{I\}$ is truncated.

We may now easily prove the important *Goldstone linked cluster theorem* [93], namely that no unlinked terms appear in the above equations (95) and (96). The proof relies on the elementary nested commutator expansion,

$$\mathrm{e}^{-S}H\mathrm{e}^{S} = H + [H,S] + \frac{1}{2!}[[H,S],S] + \cdots \ , \tag{97}$$

and the fact that all of the components of S commute with each other, by construction, so that each element of S in the parametrisation (93) is linked directly to the Hamiltonian. The reason for this is that the only non-vanishing contributions to each of the commutators in the series (97) arise from terms where one of the single-particle operators in the second-quantised form for the Hamiltonian H meets its Hermitian adjoint in one of the partitions of any one of the cluster operators S. Such non-vanishing components of the commutators form the *elementary links*. Thus, we see that the only links that can arise are between H and an operator S; there are no links between different S operators, and hence every operator S in every nested commutator must be linked to the Hamiltonian. Thus, we have proven that $\mathrm{e}^{-S}H\mathrm{e}^{S}$ is a *fully linked operator*. Unlinked or disconnected pieces simply cannot arise, even when approximations to S are made. This linkedness feature is often emphasised by writing

$$\mathrm{e}^{-S}H\mathrm{e}^{S} \equiv \{H\mathrm{e}^{S}\}_{\mathcal{L}} \ , \ \ \{HS^{n+1}\}_{\mathcal{L}} \equiv [\{HS^{n}\}_{\mathcal{L}},S] \ ; \ n = 0,1,\cdots \ . \tag{98}$$

Furthermore, we note that if H contains only up to n-body operators (i.e., it comprises sums of products of no more than $2n$ single-particle operators),

the series (97) will exactly terminate with the term involving $2n$ cluster operators, S. For example, in the common case where $H = T + V$ comprises kinetic energy and pairwise potential terms only, the series (97) terminates at terms of fourth order in S. We note that the fact that the basic exact CCM equations are thus of finite order in the coefficients $\{s_I\}$ ensures that no further truncation of the CCM equations is needed after the operator S has itself been approximated. By contrast, were the energy E_0 to be calculated, as in a standard variational theory treatment, as an expectation value,

$$E_0 = \frac{\langle\Psi_0|H|\Psi_0\rangle}{\langle\Psi_0|\Psi_0\rangle} = \frac{\langle\Phi_0|\mathrm{e}^{S^\dagger}H\mathrm{e}^{S}|\Phi_0\rangle}{\langle\Phi_0|\mathrm{e}^{S^\dagger}\mathrm{e}^{S}|\Phi_0\rangle} \quad , \tag{99}$$

the resulting expression is generally of infinite order in the correlation operator S and its adjoint $S^\dagger$, however S is truncated. Furthermore, although the cancellation of unlinked terms between the numerator and denominator of (99) may be proven [99,100], it is by no means transparent.

Bra Ground State: Up to this point we have parametrised only the ket ground state, and we have made use of the Schrödinger equation itself to extract the energy, E_0, without the need to consider the bra state. However, in order to calculate other ground-state properties we cannot avoid the use of the bra state to form expectation values. It is precisely at this point that the CCM formalism now divides into the normal (NCCM) and extended (ECCM) schemes [67]. The crucial feature of both schemes is that corresponding ket and bra states are parametrised *independently* in a way that does *not* explicitly preserve their Hermitian-adjoint relationship to one another, and which as a consequence actually breaks it at a general (e.g., SUBn) level of truncation. The underlying reasons for this have been given by Arponen and his co-workers [67,68,101], and are explained below.

We introduce the notation $\langle\tilde{\Psi}_0|$ for the bra ground state,

$$\langle\tilde{\Psi}_0| \equiv \frac{(|\Psi_0\rangle)^\dagger}{\langle\Psi_0|\Psi_0\rangle} \equiv \frac{\langle\Psi_0|}{\langle\Psi_0|\tilde{\Psi}_0\rangle} \quad , \tag{100}$$

and its corresponding NCCM parametrisation,

$$\langle\tilde{\Psi}_0| = \langle\Phi_0|\tilde{S}\mathrm{e}^{-S} \quad ; \quad \tilde{S} = \mathbb{1} + \sum_I{}' \tilde{s}_I C_I \quad . \tag{101}$$

Equation (101) preserves the explicit normalisation,

$$\langle\tilde{\Psi}_0|\Psi_0\rangle = \langle\Phi_0|\Psi_0\rangle = \langle\Phi_0|\Phi_0\rangle = 1 \quad . \tag{102}$$

Upon taking expectation values between $\langle\tilde{\Psi}_0|$ and $|\Psi_0\rangle$ it also maintains intact the fundamental CCM similarity transformation, which, as we have seen, itself encodes the linked-cluster property. Although the operator $\tilde{S}$ formally satisfies the condition,

$$\langle\Phi_0|\tilde{S} = \frac{\langle\Phi_0|\mathrm{e}^{S^\dagger}\mathrm{e}^{S}}{\langle\Phi_0|\mathrm{e}^{S^\dagger}\mathrm{e}^{S}|\Phi_0\rangle} \quad , \tag{103}$$

we maintain $\{s_I, \tilde{s}_I\}$ as a complete set of *independent* variables. If the multiconfigurational label set $\{I\}$ is truncated, exact Hermiticity will generally be violated. The full set of amplitudes $\{s_I, \tilde{s}_I\}$ provides a *complete* NCCM parametrisation of the ground state. For example, an arbitrary operator, $A \in \mathcal{G}$ has a ground-state expectation value,

$$\bar{A} \equiv \langle A\rangle \equiv \langle\tilde{\Psi}_0|A|\Psi_0\rangle$$

$$= \langle\Phi_0|\tilde{S}\mathrm{e}^{-S}A\mathrm{e}^{S}|\Phi_0\rangle = \bar{A}(s_I, \tilde{s}_I) \quad . \tag{104}$$

We note that $\bar{A}$ is again fully linked (and hence scales properly with particle number, N), even though the operator $\tilde{S}$ itself contains unlinked pieces.

The bra-state coefficients $\{\tilde{s}_I\}$ are obtained via the ground-state Schrödinger equation written in the form,

$$\langle\tilde{\Psi}_0|H = E_0\langle\tilde{\Psi}_0| \implies \langle\Phi_0|\tilde{S}(\mathrm{e}^{-S}H\mathrm{e}^{S} - E_0) = 0 \quad . \tag{105}$$

Projection onto the states $C_I^\dagger|\Phi_0\rangle$ yields the equations,

$$\langle\Phi_0|\tilde{S}(\mathrm{e}^{-S}H\mathrm{e}^{S} - E_0)C_I^\dagger|\Phi_0\rangle = 0 \;\; ; \;\; \forall I \neq 0 \quad . \tag{106a}$$

Alternatively, we can eliminate E_0 from (106a) by making use of the ket-state equation (94) projected with the state $\langle\Phi_0|\tilde{S}C_I^\dagger$, to yield the equations,

$$\langle\Phi_0|\tilde{S}\mathrm{e}^{-S}[H, C_I^\dagger]\mathrm{e}^{S}|\Phi_0\rangle = 0 \;\; ; \;\; \forall I \neq 0 \quad , \tag{106b}$$

where we have made use of (14) and (94) so that $[S, C_I^\dagger] = 0, \forall I$. Equations (106*a*,*b*) are equivalent linear sets of equations for the NCCM coefficients $\{\tilde{s}_I\}$, and use the $\{s_I\}$ coefficients as input. Formally, we may also solve (105) or, equivalently, (106*a*) or (106*b*), for the quantity $\langle\Phi_0|\tilde{S}$. It is not difficult to prove the relation,

$$\langle\Phi_0|\tilde{S} = \langle\Phi_0| + \langle\Phi_0|\mathrm{e}^{-S}H\mathrm{e}^{S}Q(E_0 - Q\mathrm{e}^{-S}H\mathrm{e}^{S}Q)^{-1}Q \quad , \tag{107}$$

where, as in (9), the operator $Q \equiv \mathbb{1} - |\Phi_0\rangle\langle\Phi_0|$ is the projector out of our model space $\mathcal{M}$, which is now spanned by the single state $|\Phi_0\rangle$.

Hellmann–Feynman Theorem: We are now in a position to show that the NCCM bra-state parametrisation has the very important property that it is *derivable from the Hellmann–Feynman theorem* [102], in a sense which we will make clear below. The Hellmann–Feynman theorem, which is extremely simple to prove, states that if we perturb our Hamiltonian, $H \rightarrow H' = H + \lambda A$, where λ is an (infinitesimally) small quantity, such that the ground-state energy changes as $E_0 \rightarrow E_0' = E_0 + \lambda \mathrm{d}E_0/\mathrm{d}\lambda + O(\lambda^2)$, then

$$\mathrm{d}E_0/\mathrm{d}\lambda \equiv \bar{A} = \langle \Psi_0 | \mathrm{d}H/\mathrm{d}\lambda | \Psi_0 \rangle \ . \tag{108}$$

Now, instead of using the expectation value functional $\bar{H}$ to calculate E_0, let us use the functional

$$\mathcal{I}_H[S] \equiv \langle \Phi_0 | \mathrm{e}^{-S} H \mathrm{e}^{S} | \Phi_0 \rangle \ . \tag{109}$$

Equations (95) and (96) show that $\mathcal{I}_H[S] = E_0$ when S is calculated from the CCM equations (96). Let us now demand that the expectation value, $\bar{A}$, of the perturbing operator is calculated from $\mathcal{I}_H[S]$ by using the Hellmann–Feynman theorem. By making use of the relations $[S, C_I^\dagger] = 0$, and $\langle \Phi_0 | C_I^\dagger = 0$, we readily prove that

$$\begin{aligned} \bar{A} &= \frac{\mathrm{d}}{\mathrm{d}\lambda} \langle \Phi_0 | \mathrm{e}^{-S} (H + \lambda A) \mathrm{e}^{S} | \Phi_0 \rangle \\ &= \langle \Phi_0 | \mathrm{e}^{-S} A \mathrm{e}^{S} | \Phi_0 \rangle + \sum_I{}' \langle \Phi_0 | \mathrm{e}^{-S} H \mathrm{e}^{S} C_I^\dagger | \Phi_0 \rangle \frac{\delta s_I}{\delta \lambda} \ . \end{aligned} \tag{110}$$

Next, we calculate δs_I from the perturbed CCM equations (96) in which $H \rightarrow H + \lambda A$, $S \rightarrow S + \delta S$. By retaining only $O(\lambda, \delta S)$ terms, and using (96) to eliminate the unperturbed term, we readily find

$$\sum_J{}' \langle \Phi_0 | C_I (E_0 - \mathrm{e}^{-S} H \mathrm{e}^{S}) C_J^\dagger | \Phi_0 \rangle \frac{\delta s_J}{\delta \lambda} = \langle \Phi_0 | C_I \mathrm{e}^{-S} A \mathrm{e}^{S} | \Phi_0 \rangle \ ; \quad \forall I \neq 0 \ , \tag{111a}$$

where we have also made use of (95). By making use of the closure relation (18), we readily find that (111a) may be written in the equivalent form,

$$\sum_J{}' C_J^\dagger | \Phi_0 \rangle \frac{\delta s_J}{\delta \lambda} = Q (E_0 - Q \mathrm{e}^{-S} H \mathrm{e}^{S} Q)^{-1} Q \mathrm{e}^{-S} A \mathrm{e}^{S} | \Phi_0 \rangle \ . \tag{111b}$$

The use of (111b) in (110) then shows that $\bar{A}$ takes precisely the previously given form, $\bar{A} = \langle \Phi_0 | \tilde{S} \mathrm{e}^{-S} A \mathrm{e}^{S} | \Phi_0 \rangle$ with the operator $\tilde{S}$ as specified in (107).

Now, it is straightforward to show that each of equations (105) and (107) implies the other. Thus, from the above derivations we have shown that the use of the Hellmann–Feynman theorem with respect to the use of the functional $\mathcal{I}_H[S]$ to calculate E_0, and the use of the CCM equations (96) to calculate the linked-cluster correlation coefficients $\{s_I\}$, implies the NCCM bra-state parametrisation (101). Conversely, we note that no other bra-state parametrisation is compatible with the Hellmann–Feynman theorem in this

way. In particular, using Hermiticity to relate $\langle\tilde{\Psi}_0|$ to $|\Psi_0\rangle$ will violate the theorem at anything less than an exact calculation. A different method of calculating ground-state expectation values $\bar{A}$ within the CCM by Kümmel [103] also violates the theorem.

Thouless [96] has pointed out that the Hellmann–Feynman theorem immediately implies that an expectation value $\bar{A}$ of an arbitrary operator A may be calculated diagrammatically from the same set of Goldstone diagrams as for the energy, $\bar{H}$, but in which each interaction potential is replaced in turn, one at a time, by the operator A. We shall see below that not only is the CCM parametrisation $\{S, \tilde{S}\}$ derivable from the Hellmann–Feynman theorem in the above sense, but all truncations are also consistent with it. Thus, if the CCM energy is calculated from $\bar{H}$ as defined in (104), then if the coefficients $\{s_I, \tilde{s}_I\}$ are truncated by retaining only some subset of them, and the resulting expression for $\bar{H}$ is expressed as the sum of some definite subset of Goldstone diagrams, then the average value $\bar{A}$ of an arbitrary operator, calculated by (104), corresponds to precisely the same subset of diagrams when each interaction is replaced in turn by the operator A.

We note that Monkhorst [104] was the first to formulate a CCM prescription for ground-state average values, $\bar{A}$, which is compatible with the Hellmann–Feynman theorem, by employing techniques of linear and higher-order response theory. It is interesting to note, however, that he never explicitly introduced a functional form for $\bar{A}$. Once supplemented [105] by the basis set effects that are needed for the practical evaluation of energy derivatives within quantum chemistry, the scheme of Monkhorst was successfully applied [106] to the prediction of vibrational spectra and to the location of transition states for decomposition reactions.

Static Variational Principle: When Arponen introduced the NCCM [67], it was basically a generalisation of the method of Monkhorst [104]. Importantly, he showed that the NCCM equations (96) and (106) for the coefficients $\{s_I, \tilde{s}_I\}$ can also be derived by requiring that the expectation value $\bar{H}$, defined as in (104), be stationary (i.e., $\delta\bar{H} = 0$) with respect to all variations in each of the *independent* variables $\{s_I, \tilde{s}_I\}$. We thus see very simply that the requirements $\delta\bar{H}/\delta\tilde{s}_I = 0$ and $\delta\bar{H}/\delta s_I = 0$ immediately yield the former equations (96) and (106*b*), respectively. We also note that at the stationary point, determined by the solutions to these NCCM equations, $\bar{H} \rightarrow \bar{H}|_{\text{stat}} = E_0$, where E_0 is given by (95), since at this stationary point equations (96) are fulfilled. The reader should beware, however, that although E_0 is thus derived from a variational (or, better, a bivariational) principle, the estimate obtained for it using a truncated set of amplitudes $\{s_I, \tilde{s}_I\}$ does *not* yield an upper bound, due to the loss of the Hermitian-adjoint relationship between the ground bra and ket states.

We may now also return to the Hellmann–Feynman theorem. If we perturb the Hamiltonian, $H \rightarrow H' = H + \lambda A$, as above, so that the new stationary

point of $\bar{H}'$ is with NCCM parameters $\{s_I + \delta s_I, \tilde{s}_I + \delta\tilde{s}_I\}$, we readily see that at the perturbed stationary point,

$$\bar{H}'|_{\rm stat} \approx \bar{H}|^0_{\rm stat} + \sum_I{}' \left(\frac{\delta\bar{H}}{\delta s_I}\delta s_I + \frac{\delta\bar{H}}{\delta\tilde{s}_I}\delta\tilde{s}_I \right)\Bigg|^0_{\rm stat} + \lambda\bar{A}|^0_{\rm stat} \ , \tag{112}$$

correct to first-order changes, and where $\bar{A}|^0_{\rm stat}$ now denotes the value of $\bar{A}$ at the unperturbed stationary point. Hence, since $\delta\bar{H}/\delta s_I = 0 = \delta\bar{H}/\delta\tilde{s}_I$ at the unperturbed stationary point, we immediately have that

$$\frac{\rm d}{{\rm d}\lambda}\langle H + \lambda A\rangle|_{\rm stat} = \frac{{\rm d}E_0}{{\rm d}\lambda} = \bar{A}|^0_{\rm stat} \ , \tag{113}$$

which is just the Hellmann–Feynman theorem. We see that the NCCM is thus fully consistent with (and derivable from) the Hellmann–Feynman theorem at any level of approximation by truncation to a subset of configurations $\{I\}$.

Near the stationary point of $\bar{H}$ we have, correct to terms of second order,

$$\bar{H} \approx E_0 + \sum_{I,J}{}' \left(\delta\tilde{s}_I E_{IJ}\delta s_J + \frac{1}{2}\delta s_I F_{IJ}\delta s_J \right) \ , \tag{114}$$

where

$$\begin{aligned} E_{IJ} &\equiv \delta^2\bar{H}/\delta\tilde{s}_I\delta s_J|_{\rm stat} \equiv (E^T)_{JI} \ , \\ F_{IJ} &\equiv \delta^2\bar{H}/\delta s_I\delta s_J|_{\rm stat} = F_{JI} \ . \end{aligned} \tag{115}$$

We note that there are no terms bilinear in the quantities $\{\delta\tilde{s}_I\}$ in (114), due to the linear way in which $\tilde{S}$ enters the expectation value (104). We may conveniently express (114) in the equivalent block-matrix form,

$$\bar{H} = E_0 + \frac{1}{2}(\delta x)^T\mathcal{H}\delta x + O(\delta x)^3 \ , \tag{116}$$

where

$$\delta x \equiv \begin{pmatrix} \delta s_I \\ \delta\tilde{s}_I \end{pmatrix} \ ; \quad \mathcal{H} \equiv \begin{pmatrix} F & E^T \\ E & 0 \end{pmatrix} = \mathcal{H}^T \ . \tag{117}$$

Linear Response to Static Perturbations: Many important physical parameters for real materials, such as polarisabilities and susceptibilities, cannot be calculated as ground-state expectation values of time-independent operators. An important tool in such cases is often *linear response theory*. Let us first consider *static linear response*. As above we subject the system Hamiltonian, to a small perturbation, $H \rightarrow H' = H + \lambda A$, and measure the consequent first-order change to the ground-state expectation value of another operator B,

$$\bar{B} \rightarrow \bar{B}' \equiv \bar{B} - \lambda R_{BA} + O(\lambda^2) \ , \tag{118}$$

where, by definition, R_{BA} is the associated *static response function.* Expanding about the stationary point of H, we have, correct to second order,

$$\bar{H}' \approx \bar{H}'|^0_{\text{stat}} + \frac{1}{2}(\delta x)^T \mathcal{H} \delta x + \lambda (\delta x)^T \frac{\delta \bar{A}}{\delta x} \quad , \tag{119}$$

where $\delta \bar{A}/\delta x$ is the column vector formed from elements $\delta \bar{A}/\delta s_I$ and $\delta \bar{A}/\delta \tilde{s}_I$ evaluated at the stationary point of $\bar{H}$ alone. The stationarity condition, $\delta \bar{H}'/\delta x = 0$, for the perturbed Hamiltonian, then gives

$$\delta x = -\lambda \mathcal{H}^{-1} \left(\frac{\delta \bar{A}}{\delta x} \right) \quad , \tag{120}$$

by making use of (119).

The average value of the operator B in the perturbed state is given by

$$\bar{B}(s_I + \delta s_I, \tilde{s}_I + \delta \tilde{s}_I) = \bar{B}(s_I, \tilde{s}_I) + \left(\frac{\delta \bar{B}}{\delta x} \right)^T \delta x + O(\lambda^2) \quad . \tag{121}$$

Comparison of (118) and (121), together with (120), yields the result,

$$R_{BA} = \left(\frac{\delta \bar{B}}{\delta x} \right)^T \mathcal{H}^{-1} \left(\frac{\delta \bar{A}}{\delta x} \right) \quad . \tag{122a}$$

By making use of the fact that $\mathcal{H}$ is a symmetric matrix, (122a) implies the symmetry relation $R_{BA} = R_{AB}$. Equation (122a) may also more explicitly be written as

$$R_{BA} = \sum_{I,J}{}' \left\{ (E^{-1})_{IJ} \left(\frac{\delta \bar{A}}{\delta s_I} \frac{\delta \bar{B}}{\delta \tilde{s}_J} + \frac{\delta \bar{B}}{\delta s_I} \frac{\delta \bar{A}}{\delta \tilde{s}_J} \right) - [(E^{-1})^T F E^{-1}]_{IJ} \frac{\delta \bar{A}}{\delta \tilde{s}_I} \frac{\delta \bar{B}}{\delta \tilde{s}_J} \right\} \quad . \tag{122b}$$

We note that the perturbation λA often destroys a possible symmetry of the Hamiltonian. An example is the calculation of the density-density response function for a translationally-invariant system. In such cases the operators $\{S, \tilde{S}\}$ must be parametrised to allow for *symmetry breaking.* Thus, even though in the symmetric ground state the stationary values of those symmetry-breaking parameters will be zero, the second-order derivatives contained in $\mathcal{H}$ of $\bar{H}$ with respect to those parameters will generally not be zero. We also note that if the matrix $\mathcal{H}$ has an eigenvalue close to zero, the linear response to the perturbation A will be large. The approach of an eigenvalue to zero (i.e., a mode becoming soft), as some internal or external parameter is varied, often signifies the approach of a *phase transition* in the system, typically to a state which differs profoundly from the (naive vacuum or) current model state, $|\Phi_0\rangle$.

Dynamics and Dynamic Variational Principle: All of the above concepts relating to the stationary Schrödinger equation and static response may now be generalised to the dynamic (i.e., time-dependent) case. We define an action-like functional, $\mathcal{A}$,

$$\mathcal{A} = \mathcal{A}[|\Psi(t)\rangle; \langle\tilde{\Psi}(t)|] \equiv \int_{-\infty}^{\infty} \mathrm{d}t \, \langle\tilde{\Psi}(t)|(\mathrm{i}\partial/\partial t - H)|\Psi(t)\rangle \ . \tag{123}$$

The requirement that $\mathcal{A}$ be stationary, $\delta\mathcal{A} = 0$, with respect to arbitrary changes in $|\Psi(t)\rangle$ and $\langle\tilde{\Psi}(t)|$ independently is completely equivalent to the dynamic Schrödinger equations,

$$\delta\mathcal{A}/\delta\langle\tilde{\Psi}| = 0 \ \Rightarrow \ \mathrm{i}(\partial/\partial t)|\Psi(t)\rangle = H|\Psi(t)\rangle \ , \tag{124a}$$

$$\delta\mathcal{A}/\delta|\Psi\rangle = 0 \ \Rightarrow \ -\mathrm{i}(\partial/\partial t)\langle\tilde{\Psi}(t)| = \langle\tilde{\Psi}(t)|H \ . \tag{124b}$$

The time-dependent states are now parametrised in the NCCM, by complete analogy with their static counterparts in (93) and (101), as follows,

$$|\Psi(t)\rangle = \mathrm{e}^{k(t)}\mathrm{e}^{S(t)}|\Phi_0\rangle \ ; \ S(t) = \sum_I{}' s_I(t)C_I^\dagger \ , \tag{125a}$$

$$\langle\tilde{\Psi}(t)| = \mathrm{e}^{-k(t)}\langle\Phi_0|\tilde{S}(t)\mathrm{e}^{-S(t)} \ ; \ \tilde{S}(t) = \mathbb{1} + \sum_I{}' \tilde{s}_I(t)C_I \ . \tag{125b}$$

The c-number $k(t)$ is a necessary scale factor to describe the time-dependence of the amplitude of the wave functions, which remain manifestly normalised to each other at all times, however,

$$\langle\tilde{\Psi}(t)|\Psi(t)\rangle = \langle\Phi_0|\Phi_0\rangle = 1 \ , \ \forall t \ . \tag{126}$$

By making use of the relation (17) we may thus express $\mathcal{A}$ in the NCCM form,

$$\mathcal{A} = \int_{-\infty}^{\infty} \mathrm{d}t \left\{ \mathrm{i}\sum_I{}' \tilde{s}_I\dot{s}_I - \bar{H}(s_I, \tilde{s}_I) \right\} \ , \tag{127}$$

where $\dot{s}_I \equiv \mathrm{d}s_I/\mathrm{d}t$, and where $\bar{H}(s_I, \tilde{s}_I)$ is as specified in (104). Stationarity of $\mathcal{A}$ with respect to the complete set of NCCM parameters $\{s_I, \tilde{s}_I\}$ then re-expresses the *dynamic variational principle*, $\delta\mathcal{A} = 0$, in the form,

$$\frac{\delta\mathcal{A}}{\delta\tilde{s}_I} = 0 \ \Rightarrow \ \mathrm{i}\dot{s}_I = \frac{\delta\bar{H}}{\delta\tilde{s}_I} \ , \tag{128a}$$

$$\frac{\delta\mathcal{A}}{\delta s_I} = 0 \ \Rightarrow \ -\mathrm{i}\dot{\tilde{s}}_I = \frac{\delta\bar{H}}{\delta s_I} \ . \tag{128b}$$

We note that although the NCCM operators S and $\tilde{S}$ have seemingly entered the parametrisations (93) and (101), or (125a, b), in a very asymmetric fashion, their fundamental dynamics is now revealed in (128a, b) to be that of a *canonically conjugate pair*. Thus, $(s_I, \tilde{s}_I)$ are revealed to be canonically

conjugate to each other in the usual terminology of classical Hamiltonian mechanics, in terms of a (classical) Hamiltonian functional $\bar{H} = \bar{H}(s_I, \tilde{s}_I)$. This conjugacy can be made absolutely precise by defining *generalised multi-configurational fields*, ϕ_I, and their *canonically conjugate multiconfigurational momenta*, π_I, as follows

$$\phi_I \equiv 2^{-\frac{1}{2}}(\tilde{s}_I + s_I) \;\; ; \quad \pi_I \equiv 2^{-\frac{1}{2}}\mathrm{i}(\tilde{s}_I - s_I) \;\; , \tag{129}$$

in terms of which the NCCM equations of motion (128a,b) can be written in the equivalent form,

$$\dot{\phi}_I = \delta\bar{H}/\delta\pi_I \;\; ; \quad \dot{\pi}_I = -\delta\bar{H}/\delta\phi_I \;\; , \tag{130}$$

which are precisely those of the classical Hamiltonian mechanics of a system described by a Hamiltonian $\bar{H} = \bar{H}(\phi_I, \pi_I)$.

By considering the dynamics of wave functions we have thus uncovered the (hitherto hidden) deep and fascinating result that the NCCM parametrisation represents an **exact** mapping of the general quantum many-body or field theory, described in terms of the complete set $\{|\Phi_0\rangle; C_I^\dagger\}$, onto the classical mechanics of a set of multiconfigurational classical fields $\{s_I, \tilde{s}_I\}$ interacting via the Hamiltonian, $\bar{H}(s_I, \tilde{s}_I) \equiv \langle\Phi_0|\tilde{S}\mathrm{e}^{-S}H\mathrm{e}^{S}|\Phi_0\rangle$. This classical Hamiltonian structure is now explored further below by considering the time-evolution of the expectation value of an arbitrary operator $A \in \mathcal{G}$.

Equation of Motion of an Arbitrary Expectation Value: We consider an arbitrary operator A with an expectation value, $\langle A\rangle = \bar{A} = \bar{A}(s_I, \tilde{s}_I; t)$, defined as in (104) but with the operators $S = S(t)$ and $\tilde{S} = \tilde{S}(t)$ now time-dependent. By making use of both the usual chain rule for partial differentiation and the equations of motion (128a,b), we readily prove that

$$\mathrm{d}\langle A\rangle/\mathrm{d}t = \langle\partial A/\partial t\rangle + \{\bar{A}, \bar{H}\} \;\; , \tag{131}$$

where the term $\langle\partial A/\partial t\rangle$ arises only from any intrinsic time-dependence of $A = A(t)$, and where we define a *generalised Poisson bracket*, $\{\bar{A}, \bar{B}\}$, as

$$\begin{aligned}\{\bar{A}, \bar{B}\} &= \frac{1}{\mathrm{i}}{\sum_I}' \left(\frac{\delta\bar{A}}{\delta s_I}\frac{\delta\bar{B}}{\delta\tilde{s}_I} - \frac{\delta\bar{B}}{\delta s_I}\frac{\delta\bar{A}}{\delta\tilde{s}_I}\right) \\ &= {\sum_I}' \left(\frac{\delta\bar{A}}{\delta\phi_I}\frac{\delta\bar{B}}{\delta\pi_I} - \frac{\delta\bar{B}}{\delta\phi_I}\frac{\delta\bar{A}}{\delta\pi_I}\right) \;\; .\end{aligned} \tag{132}$$

Thus, (131) represents the expected classical equation of motion in the canonical Hamiltonian formalism. We may complete the exploration of the classical Hamiltonian structure by considering the mapping of expectation values of commutators of operators in $\mathcal{G}$. We show below how these map onto precisely the generalised Poisson brackets (132) introduced above.

Expectation Values of Commutators: The NCCM expectation value (104) of the product of two operators may be written in the following form, by making use of the completeness relation (18),

$$\langle\tilde{\Psi}_0|AB|\Psi_0\rangle \equiv \overline{AB} = \sum_I \langle\Phi_0|\tilde{S}\mathrm{e}^{-S}A\mathrm{e}^{S}C_I^\dagger|\Phi_0\rangle\langle\Phi_0|C_I\mathrm{e}^{-S}B\mathrm{e}^{S}|\Phi_0\rangle \ . \quad (133)$$

By making use again of the definition (104) for expectation values, we readily prove the relations,

$$\delta\bar{A}/\delta s_I = \langle\Phi_0|\tilde{S}\mathrm{e}^{-S}(AC_I^\dagger - C_I^\dagger A)\mathrm{e}^{S}|\Phi_0\rangle \ ; \quad \forall I \neq 0 \ , \quad (134a)$$

$$\delta\bar{B}/\delta\tilde{s}_I = \langle\Phi_0|C_I\mathrm{e}^{-S}B\mathrm{e}^{S}|\Phi_0\rangle \ ; \quad \forall I \neq 0 \ . \quad (134b)$$

Equations (134*a*,*b*) may be used to rewrite (133) as follows,

$$\overline{AB} = \sum_I{}' \frac{\delta\bar{A}}{\delta s_I}\frac{\delta\bar{B}}{\delta\tilde{s}_I} + \sum_I \langle\Phi_0|\tilde{S}C_I^\dagger\mathrm{e}^{-S}A\mathrm{e}^{S}|\Phi_0\rangle\langle\Phi_0|C_I\mathrm{e}^{-S}B\mathrm{e}^{S}|\Phi_0\rangle \ . \quad (135)$$

By a judicious insertion of the completeness relation (18), we may rewrite (135) in the form

$$\overline{AB} = \sum_I{}' \frac{\delta\bar{A}}{\delta s_I}\frac{\delta\bar{B}}{\delta\tilde{s}_I} + \sum_{I,J}\langle\Phi_0|\tilde{S}C_I^\dagger C_J^\dagger|\Phi_0\rangle\langle\Phi_0|C_J\mathrm{e}^{-S}A\mathrm{e}^{S}|\Phi_0\rangle$$

$$\times\langle\Phi_0|C_I\mathrm{e}^{-S}B\mathrm{e}^{S}|\Phi_0\rangle \ . \quad (136)$$

We have rewritten the last term in (136) to show explicitly that it is symmetric under the interchange $A \rightleftharpoons B$, by making use of the commutation relation (14). Equation (136) thus yields the very important relation,

$$\langle\tilde{\Psi}_0|[A,B]|\Psi_0\rangle \equiv \overline{[A,B]} = \mathrm{i}\{\bar{A},\bar{B}\} \ . \quad (137)$$

In particular, we see that (131) and (137) are completely consistent with the usual quantum-mechanical Heisenberg equation of motion for an arbitrary operator $A \in \mathcal{G}$,

$$\frac{\mathrm{d}A}{\mathrm{d}t} = \frac{\partial A}{\partial t} + \frac{1}{\mathrm{i}}[A,H] \ . \quad (138)$$

Dynamic Linear Response: Let us now consider perturbing the ground state of our stationary system by a weak time-dependent perturbation, $H \to H'(t) = H + \lambda(t)A$, under which our ground-state NCCM correlation operators are perturbed as $S \to S'(t) = S + \delta S(t)$ and $\tilde{S} \to \tilde{S}'(t) = \tilde{S} + \delta\tilde{S}(t)$. Once again, we will work to leading order in λ and the associated small changes. If we now thus expand the equations of motion (128a,b) about the unperturbed ground-state stationary point, at which $\delta\bar{H}/\delta s_I = 0 = \delta\bar{H}/\delta\tilde{s}_I$, we readily find, in the same block-matrix notation introduced previously,

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix}\delta s_I \\ -\delta\tilde{s}_I\end{pmatrix} \approx \sum_J{}' \begin{pmatrix}E_{IJ} & 0 \\ F_{IJ} & E_{JI}\end{pmatrix}\begin{pmatrix}\delta s_J \\ \delta\tilde{s}_J\end{pmatrix} + \lambda(t)\begin{pmatrix}\delta\bar{A}/\delta\tilde{s}_I \\ \delta\bar{A}/\delta s_I\end{pmatrix} \quad , \tag{139a}$$

where E_{IJ} and F_{IJ} are as defined in (115). In terms of the block column matrix δx and the block matrix $\mathcal{H}$ introduced in (117) above, we may rewrite (139a) in the equivalent form,

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\delta x \approx J\mathcal{H}\delta x + \lambda(t)J\begin{pmatrix}\delta\bar{A} \\ \delta x\end{pmatrix} \quad , \tag{139b}$$

where we have introduced the block matrix, J,

$$J \equiv \begin{pmatrix}0 & \mathbb{1} \\ -\mathbb{1} & 0\end{pmatrix} \quad . \tag{140}$$

In the linear response approximation, (139b) is thus a simple first-order differential equation for the column matrix $\delta x = \delta x(t)$. It is readily seen to have the following solution,

$$\delta x = -\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t'\, \mathrm{e}^{-\mathrm{i}(t-t')J\mathcal{H}}\lambda(t')J\begin{pmatrix}\delta\bar{A} \\ \delta x\end{pmatrix} \quad , \tag{141}$$

where we have assumed $\lambda(t) \to 0$ as $t \to -\infty$.

As in the static case, to leading order, the change in expectation value $\bar{B}$ of any operator B, due to the perturbation $\lambda(t)A$, is as given by (121). The analogue of (118) for the dynamic case is now

$$\bar{B} \to \bar{B}'(t) \equiv \bar{B} - \int_{-\infty}^{t} \mathrm{d}t'\, R_{BA}(t-t')\lambda(t') + O(\lambda^2) \quad , \tag{142}$$

where $R_{BA}(t)$ is the *dynamic response function.* A comparison of (141) and (142) gives the explicit solution,

$$R_{BA}(t) = \mathrm{i}\begin{pmatrix}\delta\bar{B} \\ \delta x\end{pmatrix}^T \mathrm{e}^{-\mathrm{i}tJ\mathcal{H}}J\begin{pmatrix}\delta\bar{A} \\ \delta x\end{pmatrix}\theta(t) \quad , \tag{143}$$

where $\theta(x)$ is the usual unit-step function, equal to one if $x > 0$ and zero if $x < 0$. Equivalently, in the frequency domain, we may define the Fourier transform,

$$R_{BA}(\omega) = \int_0^\infty \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\omega t} R_{BA}(t) \ . \tag{144}$$

So long as the *dynamic matrix*, H_D,

$$H_D \equiv J\mathcal{H} \ , \tag{145}$$

has no eigenvalues with a positive imaginary part (and see below), we may insert into (144) a factor $\mathrm{e}^{-\epsilon t}$ (or, equivalently, let $\omega \to \omega + i\epsilon$) with $\epsilon > 0$ a positive infinitesimal, to ensure convergence as $t \to \infty$, and thereby obtain the result

$$R_{BA}(\omega) = \left(\frac{\delta \bar{B}}{\delta x}\right)^T (J\mathcal{H} - \omega - \mathrm{i}\epsilon)^{-1} J \left(\frac{\delta \bar{A}}{\delta x}\right) \ . \tag{146}$$

A direct comparison of (122*a*) and (146) shows that $R_{BA}(\omega = 0) = R_{BA}$, viz., that the static limit is regained, as expected, in the limit of zero frequency. One may also show from (146) that $R_{BA}(\omega) = R_{AB}(\omega)$, although it is not entirely trivial to do so.

In view of the comment below (145), it is also interesting to note that the eigenvalues of the dynamic matrix H_D come in pairs, $\pm\omega_n$, as we now show. Since H_D is not Hermitian, we may solve either for the right or left eigenvectors, ζ_n or $\tilde{\zeta}_n$, respectively,

$$H_D \zeta_n = \omega_n \zeta_n \ ; \quad \tilde{\zeta}_n^T H_D = \omega_n \tilde{\zeta}_n^T \ . \tag{147}$$

By taking the transpose of the latter (left eigenvector) equation, and by premultiplying by the matrix J, we observe that

$$-J\mathcal{H}J\tilde{\zeta}_n = \omega_n J\tilde{\zeta}_n \ \Rightarrow \ H_D(J\tilde{\zeta}_n) = -\omega_n (J\tilde{\zeta}_n) \ , \tag{148}$$

where we have used the relations, $\mathcal{H}^T = \mathcal{H}$ and $J^T = -J$. Equation (148) thus shows that $J\tilde{\zeta}_n$ is a right eigenfunction with eigenvalue $-\omega_n$ if $\tilde{\zeta}_n^T$ is a left eigenfunction with eigenvalue $+\omega_n$. Our proof is thus complete. We note, furthermore, that the stationary point of $\bar{H}$ is therefore stable only if the dynamic matrix H_D has real eigenvalues. The appearance of complex eigenvalues as some internal or external parameter is varied through some critical value denotes the onset of an instability leading to a phase transition.

We note that such dynamic response functions, $R_{BA}(t)$, are extremely important in quantum many-body or quantum field theory. For example, for bosonic field theory, if A and B are single-particle field operators, $R_{BA}(t)$ becomes the *retarded single-particle Green's function*, from which one may also easily calculate the corresponding causal Green's function.

Excited States as Normal Modes of H_D: If we now consider the dynamics of the general system near its stationary ground state, $\delta\bar{H} = 0$, in the case where there is no external perturbation, small perturbations are described by (140) with $\lambda(t) \equiv 0$,

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\delta x = H_D \delta x \quad . \tag{149}$$

Just as in classical mechanics, we now seek *normal mode* solutions, $\delta x \rightarrow \zeta_n(\omega, t) = \zeta_n \mathrm{e}^{-\mathrm{i}\omega_n t}$, where ω_n and ζ_n are given by the eigenvalue equation (147). In more explicit block-matrix form, (147) may be written as follows,

$$\begin{pmatrix} E & 0 \\ -F & -E^T \end{pmatrix} \begin{pmatrix} \delta s_I \\ \delta \tilde{s}_I \end{pmatrix} = \omega_n \begin{pmatrix} \delta s_I \\ \delta \tilde{s}_I \end{pmatrix} \quad . \tag{150}$$

Thus, the (positive) eigenvalues ω_n, which correspond to the excitation energies with respect to the ground-state energy, E_0, may be expressed purely in terms of the linked-cluster amplitudes $\{\delta s_I\}$,

$$\sum_J{}' \frac{\delta^2 \bar{H}}{\delta \tilde{s}_I \delta s_J} \delta s_J = \omega_n \delta s_I \quad , \tag{151}$$

where we have made explicit use of the definition (115) of the matrix elements E_{IJ}. Using the definition (104) for the NCCM average value, $\bar{H}$, in (151) yields

$$\sum_J{}' \langle \Phi_0 | C_I \mathrm{e}^{-S} [H, C_J^\dagger] \mathrm{e}^S | \Phi_0 \rangle \delta s_J = \omega_n \delta s_I \quad ; \quad \forall I \neq 0 \quad . \tag{152a}$$

By making use of the ground ket-state Schrödinger equation, (152a) may readily be rewritten in the equivalent form,

$$\sum_J{}' Q(\mathrm{e}^{-S} H \mathrm{e}^S - E_0) \delta s_J C_J^\dagger |\Phi_0\rangle = \omega_n \sum_I{}' \delta s_I C_I^\dagger |\Phi_0\rangle \quad ; \quad \forall I \neq 0 \quad , \tag{152b}$$

where, as before, $Q \equiv \mathbb{1} - |\Phi_0\rangle\langle\Phi_0|$. Equation (152$b$) shows that the excitation energies, ω_n, of the system are obtained by diagonalisation of the same matrix, $Q(\mathrm{e}^{-S} H \mathrm{e}^S - E_0)Q$, whose inverse is required in the formal construction (107) of the NCCM bra-state operator $\tilde{S}$ from its ket-state counterpart S.

Ket Excited States: An alternative, but (as we shall see) equivalent, way of discussing excited states is via the time-independent Schrödinger equation,

$$H|\Psi_l\rangle = E_l|\Psi_l\rangle \equiv (E_0 + \omega_l)|\Psi_l\rangle \quad ; \quad l \neq 0 \quad . \tag{153}$$

The standard CCM description of excited states has been given, *inter alia*, by Emrich [107]. Strictly speaking, the NCCM parametrisation (93) describes not only the ground state of (62), but also any state with the same quantum

numbers as the ground state and with nonzero overlap with the model state $|\Phi_0\rangle$. The possibility of obtaining multiple solutions to the nonlinear ground-state NCCM equations (96) has been discussed in some detail for a particular model case in Ref. [108].

For excited states (or, more generally, for states with zero overlap with $|\Phi_0\rangle$), we construct the respective ket wave functions $\{|\Psi_l\rangle\}$ in the NCCM in terms of a set of linear excitation operators $\{X^{(l)}\}$ which act on the corresponding exact ground-state ket wave function $|\Psi_0\rangle$,

$$|\Psi_l\rangle = X^{(l)}|\Psi_0\rangle = X^{(l)}\mathrm{e}^S|\Phi_0\rangle \quad . \tag{154}$$

The operator $X^{(l)}$ is again decomposed wholly in terms of creation operators, $\{C_I^\dagger;\ I \neq 0\}$, with respect to $|\Phi_0\rangle$,

$$X^{(l)} = \sum_I{}' x_I^{(l)} C_I^\dagger \quad . \tag{155}$$

Hence the operators $X^{(l)}$ and S commute; and the prime on the sum in (155) ensures that $\langle\Phi_0|\Psi_l\rangle = 0$. For extended systems with more than one phase, for example, the so-called ground-state parametrisation (93) will generally yield only the lowest state of a given symmetry imposed implicitly by the particular choice of model state. Indeed, phase transitions may be detected within this CCM description by observing, for fixed model state $|\Phi_0\rangle$, the onset of "excited" states of negative excitation energy (or, hence, "de-excited" states) from the so-called excited-state parametrisation (154)–(155), as some internal or external parameter is varied through some critical value [109].

By combining the ground- and excited-state Schrödinger equations (93) and (154), we readily find the NCCM eigenvalue equation,

$$\mathrm{e}^{-S}[H, X^{(l)}]\mathrm{e}^S|\Phi_0\rangle = \omega_l X^{(l)}|\Phi_0\rangle \quad , \tag{156a}$$

or, equivalently,

$$(\mathrm{e}^{-S}H\mathrm{e}^S - E_0)X^{(l)}|\Phi_0\rangle = \omega_l X^{(l)}|\Phi_0\rangle \quad , \tag{156b}$$

for the excitation energy, $\omega_l \equiv E_l - E_0$, directly. By taking the inner products of (156b) with each member of the set $\{C_I^\dagger|\Phi_0\rangle;\ I \neq 0\}$ we obtain a coupled set of homogeneous linear eigenvalue equations for the excited ket-state configuration coefficients $\{x_I^{(l)}\}$, which are identical to (152a). We note also that the left-hand side of (156a) may be developed as a nested commutator expansion, analogous to (97). In this way, we note that by making use of the commutativity relation, $[X^{(l)}, S] = 0$, the excited-state NCCM equations may, rather simply and very conveniently, be derived from their ground-state counterparts as follows. Thus, we replace each multinomial term in the coefficients $\{s_I\}$ arising from the expansion of the similarity transform in (94) with a corresponding set of terms in which each single coefficient s_J is replaced

one at a time in turn by the corresponding factor $x_J^{(l)}$; and we simply drop the zeroth-order (inhomogeneous) term.

We also note that by projecting (156*b*) with the state $\langle\Phi_0|\tilde{S}$, and by making use of the bra-state Schrödinger equation (105), we readily prove the following relation,

$$\omega_l\langle\Phi_0|\tilde{S}X^{(l)}|\Phi_0\rangle = 0 \ . \tag{157}$$

Equation (157) immediately yields the required orthogonality relation,

$$\langle\tilde{\Psi}_0|\Psi_l\rangle = 0 \ , \ \text{unless} \ \omega_l = 0 \ . \tag{158}$$

Clearly, as $\omega_l \rightarrow 0$, the corresponding mode becomes soft. This mode may thus be easily excited into macroscopic occupation, and the corresponding ground state can undergo a phase transition into a state of markedly different character.

Hierarchical Approximation Schemes: In order to implement any of the above elements of the single-reference version of the NCCM, we need only to choose an appropriate model state or cyclic vector $|\Phi_0\rangle$, and then to approximate the corresponding sets of equations. The standard way to do this is to truncate the otherwise complete set of multiconfigurational set-indices $\{I\}$ to some appropriate finite or infinite subset. There are many ways of doing this, and in general one must be guided by the physics. One also needs in practice to develop hierarchical approximation schemes, in which at each increasing level one systematically incorporates more many-body correlations.

One of the simplest and intuitively most appealing such systematic hierarchical approximation schemes is the SUBn scheme for the ground state, or the more general SUB(m,n) scheme for excited states in the time-independent formalism. In the latter case, all amplitudes $\{x_I^{(l)}\}$ and $\{s_I\}$ which describe clusters of more than m and n particles (or particle/hole pairs in the case of number-conserving Fermi systems) respectively, are set to zero. The remaining equations, derived as described from (94) and (156) by taking their respective inner products with the wave functions $C_I^\dagger|\Phi_0\rangle$ of the configurations retained, are then solved without further approximation (except those introduced by the computational algorithms). In the static SUB(m,n) approximation the excitation energies $\{\omega_l\}$ are equivalently obtained by diagonalisation of the operator $Q(\mathrm{e}^{-S}H\mathrm{e}^{S} - E_0)Q$, where S is now the SUBn-approximated ground-state correlation operator, within some subspace of the multiconfigurational states defined by the truncation index m.

In general, there is no particular reason, at least, from within the static CCM formalism for ground and excited states, why the truncation indices m and n should not be chosen differently. Indeed, this freedom provides an additional degree of flexibility. Further work in this connection [110] has shown that by further embedding the theory of linear response within the CCM, each of the usual energy-weighted moment sum rules for the dynamic

(liquid) structure function may, in principle, be exactly decomposed into an infinite cluster hierarchy of sub-sum-rules.

The above work [110] also provides a set of independent exact rules for the CCM cluster amplitudes, against which approximation schemes in both static ground- and excited-state formalisms can be tested for mutual compatibility. Furthermore, it provides a mechanism whereby any additional knowledge of the system (obtained either from experiment or from alternative theory or phenomenology) involving either the excitation spectrum or the ground-state correlations, may be used to extract information, from within the CCM, on the other. Finally, in this context, it is interesting to note that by making the simple approximation that the lowest members of the CCM sub-sum-rules are saturated by a single collective (or "giant resonance") state, we regain the important Bijl–Feynman relation [111] for the excitation spectrum in terms of the static structure function.

We note that for an infinite homogeneous system, translational invariance implies that the plane-wave single-particle basis is exact, and the ensuing momentum conservation then in turn implies that the one-body partition, S_1, of the correlation operator S, is identically zero. In such cases the lowest non-trivial SUBn approximation is the SUB2 scheme. This is already an extremely rich approximation. Amongst other terms it includes all of the two-body ladder diagrams and all of the two-body particle/hole ring diagrams [112].

We note that other CCM truncation schemes apart from the SUBn scheme have been used. In the case of pure hard-core potentials, for example, the SUBn scheme is not well defined, and for strongly repulsive (but not infinite hard-core) potentials the convergence is poor. A typical example of this case is the internucleon force in nuclear physics. The cure, however, is simple. It leads to the so-called (hard-core) HCSUBn scheme (otherwise known as the χ_n-truncation or Bochum truncation scheme). Thus, at a fixed ordinary SUBn level, one first identifies that subset of terms which when iterated together lead only to diagrams which are still contained in this original particular SUBn class when each bare interaction V is replaced by a ladder-summed G-matrix, and where the relative time orderings of the remaining interactions are kept fixed. The resulting HCSUBn approximation scheme [8,9,67] is closely related to the hole-line expansion of Bethe within the framework of time-independent perturbation theory.

For systems defined on a discrete spatial lattice, a localised approximation scheme called the LSUBn scheme has also been used to considerable advantage. In this scheme all possible many-body cluster configurations I are retained which occur in all different spatial locales of n contiguous sites on the lattice. Thus, to enumerate the fundamentally distinct LSUBn configurations, one must first evaluate the different *lattice animals* of size n which are inequivalent up to lattice symmetries (i.e., translations, rotations, and reflections) shared by the Hamiltonian. Then, on each such distinct lattice animal, one must consider all possible distinct sub-configurations (both con-

nected and disconnected). Such LSUBn schemes have very successfully been applied to both quantum spin-lattice models (particularly of antiferromagnetism) [113–118] and lattice gauge theories [75–81].

All of the above schemes share the feature that the cluster amplitudes relating to configurations I not retained in the approximations are set to zero. For some purposes this may be too drastic an approximation. An alternative would be to attempt to approximate such amplitudes $\{s_I, \tilde{s}_I\}$ belonging to configurations I outside the basic subset retained by a given approximation, in terms of those retained. The invention and investigation of such schemes is at the forefront of current progress in the CCM [84].

3.6 Relationship of the CCM to TIPT and Goldstone Diagrams

For the sake of the simplest comparison with time-independent perturbation theory (TIPT) we consider a many-body Hamiltonian, $H = H_0 + V$, where H_0 is a one-body operator (e.g., kinetic energy) and V is a two-body interaction term. We also work in a single-particle basis, $|\alpha\rangle \equiv c_\alpha^\dagger|0\rangle$, in which H_0 is diagonal, so that in its second-quantised form,

$$H_0 = \sum_\alpha \epsilon_\alpha c_\alpha^\dagger c_\alpha \quad , \tag{159}$$

where $\{c_\alpha^\dagger\}$ are the generic single-particle creation operators (viz., the bosonic operators $b_\alpha^\dagger$ or the fermionic operators $a_\alpha^\dagger$ of Sect. 2). In the notation introduced in Sect. 2, we also work with a model state $|\Phi_0\rangle$ and a complete set of many-body states $C_I^\dagger|\Phi_0\rangle$ which are constructed to be eigenstates of H_0,

$$H_0|\Phi_0\rangle = \mathcal{E}_0|\Phi_0\rangle \;\; ; \;\; H_0 C_I^\dagger|\Phi_0\rangle = (\mathcal{E}_0 + e_I)C_I^\dagger|\Phi_0\rangle \quad . \tag{160}$$

Examples of such sets $\{|\Phi_0\rangle; C_I^\dagger\}$ have been given in Sects. 2.1–2.3.

We note that since H_0 is diagonal in the chosen basis, each term in (159) contains exactly one destruction operator with respect to $|\Phi_0\rangle$, since either one or other (but not both) of $c_\alpha^\dagger$ or c_α will destroy $|\Phi_0\rangle$. Thus, the nested commutator expansion (97) for $H \to H_0$ will terminate at the second term on the right-hand side,

$$\mathrm{e}^{-S} H_0 \mathrm{e}^S = H_0 + [H_0, S] \quad . \tag{161}$$

By making use of (160) and (161), we readily prove the following relation,

$$\langle\Phi_0|C_I \mathrm{e}^{-S} H_0 \mathrm{e}^S|\Phi_0\rangle = \mathcal{E}_0\delta_{I0} + e_I s_I \quad , \tag{162}$$

where we have also made use of the properties (15) and (17) of the multiconfigurational creation operators $\{C_I^\dagger\}$. If we now insert (162) into the fundamental ket-state CCM equations (95) and (96), we find that the exact ground-state energy E_0 is given in terms of the unperturbed or model state energy $\mathcal{E}_0$ as

$$E_0 - \mathcal{E}_0 = \langle \Phi_0 | \mathrm{e}^{-S} V \mathrm{e}^{S} | \Phi_0 \rangle \ , \tag{163a}$$

in which the required ket-state cluster coefficients $\{s_I\}$ are given by the non-linear equations,

$$s_I = -\frac{1}{e_I} \langle \Phi_0 | C_I \mathrm{e}^{-S} V \mathrm{e}^{S} | \Phi_0 \rangle \ ; \ \forall I \neq 0 \ . \tag{163b}$$

It is now very straightforward to develop a diagrammatic representation of (163a,b) by complete analogy with the standard development of Goldstone (time-ordered) diagrams for TIPT (and see, e.g., Ref. [119]). Indeed, if one uses the nested commutator expansion (97) with $H \rightarrow V$ in (163b), and makes a straightforward iteration of the set of equations (163b) in powers of the potential V, one generates automatically the Goldstone diagrams for the linked-cluster amplitudes $\{s_I\}$. If no truncation is made, one generates in principle this way the complete set of diagrams. Alternatively, if the cluster operator S is truncated along the lines indicated above, a partial subset of diagrams is generated. Substitution of the resulting amplitudes into (163a) gives the corresponding expression for the energy in terms of a set of fully linked and closed (i.e., with no free ends) Goldstone diagrams.

We may equivalently derive (163b) and its counterpart for the bra-state cluster amplitudes $\{\tilde{s}_I\}$ from the static variational principle,

$$\frac{\delta \bar{H}}{\delta \tilde{s}_I} = 0 \ \Rightarrow \ s_I = -\frac{1}{e_I} \frac{\delta \bar{V}}{\delta \tilde{s}_I} = -\frac{1}{e_I} \langle \Phi_0 | C_I \mathrm{e}^{-S} V \mathrm{e}^{S} | \Phi_0 \rangle \ , \tag{164a}$$

$$\frac{\delta \bar{H}}{\delta s_I} = 0 \ \Rightarrow \ \tilde{s}_I = -\frac{1}{e_I} \frac{\delta \bar{V}}{\delta s_I} = -\frac{1}{e_I} \langle \Phi_0 | \tilde{S} \mathrm{e}^{-S} [V, C_I^{\dagger}] | \Phi_0 \rangle \ . \tag{164b}$$

Equations (164a,b) are precisely the *Dyson equations* for the NCCM amplitudes $\{s_I, \tilde{s}_I\}$, in which the unperturbed (or model) cluster excitation energies, e_I, appear in the familiar guise of the *energy denominators* of Rayleigh–Schrödinger perturbation theory. The iterative solutions of (164a,b) lead to a set of terms which can be put into one-to-one correspondence with classes of Goldstone diagrams. Whereas iteration of (164a) leads, as we have already discussed, automatically to a set of linked diagrams for the linked-cluster amplitudes $\{s_I\}$, iteration of (164b) is easily seen to lead to unlinked diagrams for the amplitudes $\{\tilde{s}_I\}$. Nevertheless, all expectation values $\bar{A} = \bar{A}(s_I, \tilde{s}_I)$ calculated by (104) for all such contributions from all diagrams for s_I and $\tilde{s}_I$ remain connected, as we have shown previously.

In the usual (physicist's) convention, one draws Goldstone diagrams with "time" axis running upwards (i.e., where matrix elements such as those in (164a,b) are read from right to left, and drawn, correspondingly, from bottom to top). Furthermore, by convention, $|\Phi_0\rangle$ plays the role of vacuum, so that only particle and hole lines which are excited from the model state are explicitly drawn. In this convention, the amplitude s_I is thus a "bottom amplitude" containing no legs from below and a set of particle/hole legs emanating from

above corresponding to the configuration denoted by the set-index I. Correspondingly, $\tilde{s}_I$ is a "top amplitude".

Very related to the earlier discussion of the linked-cluster nature of the bottom amplitudes $\{s_I\}$ is the concept of *generalised time ordering* (GTO). This has traditionally been a rather useful formal device for classifying and combining certain classes of Goldstone diagrams [120]. It is based on a factorisation property of Goldstone diagrams containing disjoint (i.e., separately identifiable) sets I and J of particle/hole legs with unperturbed (or model) energies e_I, as given by (160). The simple identity

$$\frac{1}{e_I(e_I+e_J)}+\frac{1}{e_J(e_I+e_J)}=\frac{1}{e_Ie_J}\quad , \tag{165}$$

may then be used to factorise the individual energy denominators as on the right-hand side of (165), after the diagrams corresponding to all permitted time orderings which preserve the nature of the subclusters (i.e., do not change particles into holes, or vice versa) are combined together. It transpires [67,68] that the linked nature of the bottom amplitudes $\{s_I\}$ implies that the NCCM automatically generates diagrams (for the energy, for example) with such a GTO property in the "downward" direction (i.e., backwards in time). Conversely, the fact that the top amplitudes $\{\tilde{s}_I\}$ are generally unlinked implies the absence of a GTO property in the "upward" direction.

The upshot is that all of the diagrams for the energy obtained by iteration of the NCCM Dyson equations, as explained above, may thus be represented by what are termed *normal GTO tree diagrams* or NCCM trees [67]. Roughly speaking these are diagrams which "branch out" in the downward direction only (– and hence which represent the root system of a real tree, rather than its visible branch structure). It has been rather fully explained elsewhere [67] how each branch (– or root) of such a generalised diagram corresponds to a set of particle/hole lines labelled by a member of the complete set $\{I\}$. If such a (downward) closed NCCM tree diagram for the energy is divided into two open portions by cutting a single branch I, the bottom part constitutes a diagram for s_I, and will always itself be linked; whereas the top part constitutes a diagram for $\tilde{s}_I$, and may be unlinked.

We note again that whereas the NCCM equations *may* be iterated to yield terms corresponding to classes of Goldstone diagrams, we essentially never actually solve the equations this way in practice. In essence, the CCM performs a massive automatic resummation (to infinite order) of many (infinite) sets of Goldstone diagrams, and this is one of the underlying strengths of the method. By solving the equations as they stand (i.e., without iteration) at any level of truncation for the set of amplitudes $\{s_I, \tilde{s}_I\}$, we anticipate (and usually find) much better convergence properties in the hierarchical truncation scheme under consideration than the perturbation series.

Clearly, once we have made a particular choice of model state and multiconfigurational creation operators, and have chosen a particular truncation scheme, the practical implementation of the NCCM breaks into two tasks: (i)

the algebraic evaluation of NCCM expectation values (104) and their functional derivatives; and (ii) the numerical solution of the resulting equations, e.g., (164*a*,*b*), for the truncated set $\{s_I, \tilde{s}_I\}$, in terms of which all physical quantities may be evaluated. We note that the underlying similarity transformation and the (usually) finite number of terms in the resulting nested commutator expansions (97), which lie at the heart of the CCM, make the first task well suited to the use of *computer-algebraic methods*. The interested reader is referred to Ref. [118], for example, for a thorough discussion of how this may be implemented very efficiently in practice for lattice Hamiltonian systems. The second task, namely the numerical solution of the equations so derived, normally presents no great difficulties, since the CCM equations are well-conditioned (unlike their CIM counterparts), and are also found in practice to be extremely robust in almost all applications.

3.7 The Multi-Reference Normal CCM

Physically, one might intuit that the single-reference version of the NCCM discussed above in Sects. 3.5 and 3.6 is most likely to be a good approach for "closed-shell" systems, for which a single model state $|\Phi_0\rangle$ suffices for a reasonable zeroth-order description. Conversely, we turn now to "open-shell" systems for which, *a priori*, a multi-reference approach appears to provide a more reasonable calculational framework. We argue by analogy with the single-reference ground-state NCCM approach discussed in Sect. 3.5. Thus, it is clear that the single-reference ground-state scheme may be viewed at a rather shallow level as simply providing, in any particular approximation scheme, a corresponding (and, evidently, a very sophisticated and clever) partial resummation of an infinite set of terms (or Goldstone diagrams) in the nondegenerate version of TIPT, as outlined in Sect. 3.6. As we have seen in Sect. 3.5, the basic CCM equations automatically embody the historically important linked cluster theorem of Goldstone [93] for the energy.

In the same spirit, Brandow [120] first showed that in the intermediate normalisation scheme there exists a similar linked diagram expansion, namely the *linked valence expansion*, in the degenerate version of TIPT. This was the first such formulation which was both size-extensive and size-consistent, where size-extensivity [52] is the property that the leading term in the energy of an N-particle system scales linearly with N, and where size-consistency [121] implies that a many-body wave function dissociates correctly into non-interacting fragments under infinite separation of the fragments. Brandow showed, in particular, how it was necessary for his formulation to be given in terms of a so-called *"complete model space."* This is defined to be one in which the multi-reference model space contained all possible N-body configurations (or Slater determinants for the usual number-conserving fermionic case to which the formalism is applied) that can be formed by distributing the valence particles among a selected set of valence orbitals. Brandow

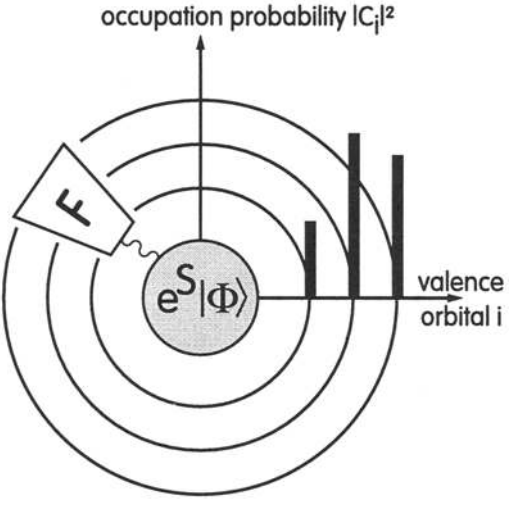

Fig. 1. A schematic representation of the essential elements of the multi-reference normal coupled cluster parametrisation of the ket-state energy eigenfunctions of an open-shell system.

further showed how his perturbative expansion could be expressed diagrammatically in terms of so-called *"folded diagrams."* Several authors have since described how to embody and extend this degenerate version of TIPT within the CCM framework, via a multi-reference approach. Perhaps the earliest within the physics (as opposed to chemistry) community were Kümmel and his co-workers [122]. Many other variants exist, most of which have been especially developed for use in quantum chemistry, but for present pedagogical purposes only this version is outlined below. Its essential ingredients are also indicated very schematically in Figure 1.

Thus, as in Sect. 3.1, let us consider for illustrative purposes, a closed-shell N-fermion system, whose CCM single-reference model state is now indicated as $|\Phi_0^N\rangle$, and whose exact ground-state energy is E_0^N. To this closed-shell system we imagine adding valence fermions (or holes) one at a time. In the notation of Sect. 2.3 we may now distinguish three sorts of single-fermion states $|\alpha_k\rangle$, namely: (i) orbitals occupied in $|\Phi_0^N\rangle$ (labelled $\alpha_k \rightarrow \nu_k$); (ii) valence orbitals (labelled $\alpha_k \rightarrow i_k$) partially occupied by the valence particles outside the core (and/or valence holes inside the core); and (iii) the remaining "unoccupied" orbitals (labelled $\alpha_k \rightarrow \rho_k$). The multi-reference CCM ansatz for the exact $(N+1)$-particle states is given as follows,

$$|\Psi_\lambda^{N+1}\rangle = \sum_{i \in \mathcal{V}} \mathrm{e}^S [1 + F^{(1)}] a_i^\dagger |\Phi_0^N\rangle C_i^\lambda \quad , \tag{166}$$

where the correlation operator S is assumed known from the N-body "closed-shell" NCCM calculation, and where the sum on the index i runs over the set

$\mathcal{V}$ of valence orbitals considered as actually degenerate or quasi-degenerate. Thus, the states $\{a_i^\dagger|\Phi_0^N\rangle;\ i \in \mathcal{V}\}$ form a set of multi-reference $(N+1)$-body Slater determinants for the set of low-lying states (labelled λ) which we are attempting to construct.

Whereas the coefficients $\{C_i^\lambda\}$ determine the mixture of uncorrelated states in the multi-reference model space, the operator $F^{(1)}$ describes the dressing of the bare valence particle by its interaction with the core. Thus, we have the decomposition,

$$F^{(1)} = \sum_{n=1}^{N+1} F_n^{(1)} \quad , \tag{167}$$

where, for example, $F_1^{(1)}$ describes the one-body (Hartree-Fock) part of the valence problem,

$$F_1^{(1)} = \sum_{\rho} \sum_{i \in \mathcal{V}} \langle \rho|F_1^{(1)}|i\rangle\, a_\rho^\dagger a_i \quad , \tag{168a}$$

and $F_2^{(1)}$ describes the *"core polarisation"* terms which arise from the correlations between the valence particle and any one core particle,

$$F_2^{(1)} = \frac{1}{(2!)^2} \sum_{\eta_1,\eta_2} \sum_{\nu} \sum_{i \in \mathcal{V}} \langle \eta_1 \eta_2|F_2^{(1)}|i\nu\rangle_A\, a_{\eta_1}^\dagger a_{\eta_2}^\dagger a_\nu a_i \quad , \tag{168b}$$

where the labels η_k denote any extra-core state (i.e., valence or "unoccupied"), and hence run over i_k and ρ_k. We note that the CCM ansatz (166) is completely general, provided only that, just as in its single-reference counterpart (66*a*), the states $|\Psi_\lambda^{N+1}\rangle$ do not have zero inner product with all of the wave functions $\{a_i^\dagger|\Phi_0^N\rangle;\ i \in \mathcal{V}\}$ which now comprise the model space $\mathcal{M}$.

The corresponding ansatz for the two valence-particle $(N+2)$-fermion wave function is given as follows,

$$|\Psi_\mu^{N+2}\rangle = \sum_{i_1,i_2 \in \mathcal{V}} \mathrm{e}^S \left[1 + F^{(1)} + \frac{1}{2} : F^{(1)^2} : + F^{(2)}\right] a_{i_1}^\dagger a_{i_2}^\dagger |\Phi_0^N\rangle C_{i_1 i_2}^\mu \quad , \tag{169}$$

where the factor of one-half in the quadratic $F^{(1)}$ term describing two "dressed" but uncorrelated valence particles is present, as usual, to avoid double-counting each excitation. We note that this term is also normal-ordered so as to avoid contractions (or links) between the two $F^{(1)}$ operators, since these are more properly contained in the genuine two-valence-particle-plus-core correlation operator, $F^{(2)}$,

$$F^{(2)} = \sum_{n=2}^{N+2} F_n^{(2)} \quad . \tag{170}$$

If we proceed further in this fashion to add an arbitrary number of valence particles outside the core, we rapidly arrive at the normal-ordered exponential ansatz (where the normal ordering is always performed with respect to

an appropriate closed-shell system or core) first written down explicitly by Lindgren [123], although the formulation of Ey [122] is completely equivalent.

By inserting the CCM parametrisations (166) and (169) into the respective $(N+1)$-body and $(N+2)$-body Schrödinger equations, and by premultiplying as usual by the factor e^{-S}, it is straightforward to derive equations for the energy eigenvalues E_λ^{N+1} and E_μ^{N+2}. Suitable projections onto the model space $\mathcal{M}$ thus lead to secular equations for the coefficients C_i^λ and $C_{i_1 i_2}^\mu$. These may be represented [8,9,122] as generalised eigenvalue equations for fully-linked one- and two-body *effective Hamiltonians,* respectively, which yield the *folded diagrams* of degenerate many-body perturbation theory. The corresponding eigenvalues are the respective excitation energies; for example, in the one-valence case, $\epsilon_\lambda \equiv E_\lambda^{N+1} - E_0^N$. Similarly, by projecting out of $\mathcal{M}$ onto "unoccupied" states, one derives the equations which determine the matrix elements of the operators $F^{(1)}$ and $F^{(2)}$. The interested reader is referred to the literature cited above, and to the article in the present volume by Kaldor, for further details.

In the original multi-reference formulation, the CCM and the associated linked diagrammatic expansions were restricted to complete model spaces and to the intermediate normalisation scheme. However, complete model spaces have prohibitively large dimensionalities, even when the number of active valence orbitals is restricted to relatively few. Mukherjee [124] first showed that for general incomplete model spaces, the condition that both the CCM cluster operators and the effective Hamiltonians are connected is normally incompatible with our previous choice of intermediate normalisation. Conversely, by abandoning this normalisation, these connectivity properties may be reinstated. A more detailed discussion of these points would, however, take us too far afield for present purposes, and the interested reader is referred to the literature [125].

3.8 Formal Elements of the Extended CCM

In the NCCM discussed so far, while all ground-state expectation values (104), $\bar{A} = \bar{A}(s_I, \tilde{s}_I)$, are linked-cluster quantities, and while the bottom amplitudes $\{s_I\}$ are also fully linked, the top amplitudes $\{\tilde{s}_I\}$ contain unlinked terms. For some purposes we might wish all of our basic amplitudes to be linked, and this is the basic motivation for the development of the *extended coupled cluster method.* (ECCM). We note firstly that the relations (14), (93), and (104) immediately imply that the NCCM top amplitudes are simply the ground-state expectation values of the multiconfigurational creation operators,

$$\tilde{s}_I = \langle C_I^\dagger \rangle \ . \tag{171}$$

If we now define the corresponding linked or connected quantities as

$$\tilde{\sigma}_I \equiv \langle C_I^\dagger \rangle_{\mathrm{linked}} \equiv \langle C_I^\dagger \rangle_{\mathrm{c}} \ , \tag{172}$$

our earlier discussion in Sect. 3.4 on general cumulant expansions immediately yields the basic ECCM parametrisation,

$$\tilde{S} = \mathrm{e}^{\tilde{\Sigma}} \;\; ; \quad \tilde{\Sigma} = \sum_I{}' \tilde{\sigma}_I C_I \;\; . \tag{173}$$

Although the cluster amplitudes $\{s_I, \tilde{\sigma}_I\}$ are a complete set of ECCM linked quantities, it is very convenient to define a new set of bottom ECCM amplitudes, $\sigma_I = \sigma_I(s_J, \tilde{\sigma}_J)$, such that the ECCM amplitudes $(\sigma_I, \tilde{\sigma}_I)$ again form a canonically conjugate pair, by complete analogy with the canonical conjugacy of the NCCM pairs $(s_I, \tilde{s}_I)$ expressed by (128*a,b*). This is very easy to achieve by writing the action-like functional (123) in the form,

$$\begin{aligned} \mathcal{A} &= \int_{-\infty}^{\infty} \mathrm{d}t \, \langle \Phi_0 | \mathrm{e}^{\tilde{\Sigma}(t)} \mathrm{e}^{-S(t)} (\mathrm{i}\partial/\partial t - H) \mathrm{e}^{S(t)} | \Phi_0 \rangle \\ &= \int_{-\infty}^{\infty} \mathrm{d}t \, \left[-\mathrm{i} \langle \Phi_0 | \dot{\tilde{\Sigma}}(t) \mathrm{e}^{\tilde{\Sigma}(t)} S(t) | \Phi_0 \rangle - \bar{H} \right] \;\; , \end{aligned} \tag{174}$$

where, in the second line of (174), we have integrated by parts. Insertion of a complete set (18) of multiconfigurational states into (174) then yields the ECCM form of the action-like functional,

$$\begin{aligned} \mathcal{A} &= \int_{-\infty}^{\infty} \mathrm{d}t \left\{ -\mathrm{i} \sum_I{}' \dot{\tilde{\sigma}}_I \sigma_I - \bar{H}(\sigma_I, \tilde{\sigma}_I) \right\} \\ &= \int_{-\infty}^{\infty} \mathrm{d}t \left\{ \mathrm{i} \sum_I{}' \tilde{\sigma}_I \dot{\sigma}_I - \bar{H}(\sigma_I, \tilde{\sigma}_I) \right\} \;\; , \end{aligned} \tag{175}$$

where the new ECCM bottom operator Σ is defined as

$$\Sigma |\Phi_0\rangle \equiv Q \mathrm{e}^{\tilde{\Sigma}} S |\Phi_0\rangle \;\; ; \quad \Sigma \equiv \sum_I{}' \sigma_I C_I^{\dagger} \;\; , \tag{176a}$$

in terms of $Q \equiv \mathbb{1} - |\Phi_0\rangle\langle\Phi_0|$, and amplitudes $\{\sigma_I\}$ given as follows,

$$\sigma_I = \langle \Phi_0 | C_I \mathrm{e}^{\tilde{\Sigma}} S | \Phi_0 \rangle \;\; \Leftrightarrow \;\; s_I = \langle \Phi_0 | C_I \mathrm{e}^{\tilde{\Sigma}} \Sigma | \Phi_0 \rangle \;\; . \tag{176b}$$

It is evident from the definition (176*b*) that the ECCM bottom amplitudes σ_I are fully linked, since S is a linked-cluster operator. Furthermore, stationarity of $\mathcal{A}$ from (175) now shows that the ECCM amplitudes $\{\sigma_I, \tilde{\sigma}_I\}$ form canonically conjugate pairs,

$$\frac{\delta \mathcal{A}}{\delta \tilde{\sigma}_I} = 0 \;\; \Rightarrow \;\; \mathrm{i}\dot{\sigma}_I = \frac{\delta \bar{H}}{\delta \tilde{\sigma}_I} \;\; , \tag{177a}$$

$$\frac{\delta \mathcal{A}}{\delta \sigma_I} = 0 \;\; \Rightarrow \;\; -\mathrm{i}\dot{\tilde{\sigma}}_I = \frac{\delta \bar{H}}{\delta \sigma_I} \;\; , \tag{177b}$$

in complete analogy with the canonical conjugacy of the NCCM pairs $\{s_I, \tilde{s}_I\}$ expressed via (128*a*,*b*).

Although we shall not go through the details here, we note that many of the NCCM results of Sect. 3.5 may now be generalised to the ECCM [67,68,101,126]. Many of the key distinguishing features of the ECCM follow from its *double* exponential structure and hence also its double similarity transform structure,

$$\begin{aligned} \bar{A} \equiv \langle \tilde{\Psi}_0 | A | \Psi_0 \rangle &= \langle \Phi_0 | \mathrm{e}^{\tilde{\Sigma}} \mathrm{e}^{-S} A \mathrm{e}^{S} | \Phi_0 \rangle \\ &= \langle \Phi_0 | \mathrm{e}^{\tilde{\Sigma}} \mathrm{e}^{-S} A \mathrm{e}^{S} \mathrm{e}^{-\tilde{\Sigma}} | \Phi_0 \rangle \\ &\equiv \bar{A}(\sigma_I, \tilde{\sigma}_I) \ . \end{aligned} \tag{178}$$

In turn, these features lead to a well-defined *double linking* property of the corresponding ECCM diagrams [67]. By contrast to the CIM and NCCM, *all* of the basic ECCM amplitudes $\{\sigma_I, \tilde{\sigma}_I\}$ that now completely characterise the many-body system are linked-cluster quantities. In turn, they all therefore obey the important *cluster property,* namely that they approach zero asymptotically as any subset of the particles described by the configuration set-index I becomes far removed from the remainder. The entire system may thus be parametrised in terms of a complete set of ECCM multilocal, classical (i.e., *c*-number) amplitudes. Just as we have shown above for the NCCM, we can also show [101] how the ECCM maps an arbitrary quantum many-body or field-theoretic system with underlying Schrödinger dynamics onto a classical field theory.

Extensions of the ECCM have been made [68,126] to consider both excited states and general dynamical behaviour. It has thus been demonstrated how the amplitudes $\{\sigma_I, \tilde{\sigma}_I\}$ may also be viewed as *generalised (many-body) mean fields* or *quasilocal order parameters,* by considering their small-amplitude oscillations around a stationary equilibrium point, just as we did for the NCCM in Sect. 3.5. The overall structure of the ECCM is indicated very schematically in Figure 2, and the interested reader is referred to the literature [67–69,90,101,126–132] for further details.

We note that, to the best of our knowledge, the ECCM is unique as a formulation of quantum many-body/field theory in which *every* fundamental amplitude exactly obeys the cluster property at all levels of approximation. It is clear that only such formulations have the possibility to describe both the *local* properties of many-body systems and such *global* properties as their phase transitions, states of topological excitation or deformation, spontaneous symmetry breaking, general nonequilibrium behaviour, and nonlinear (large-amplitude) response.

3.9 Other Aspects of Coupled Cluster Theory

The range of individual components which are now contained under the general umbrella of coupled cluster theory or the CCM is large and varied, as

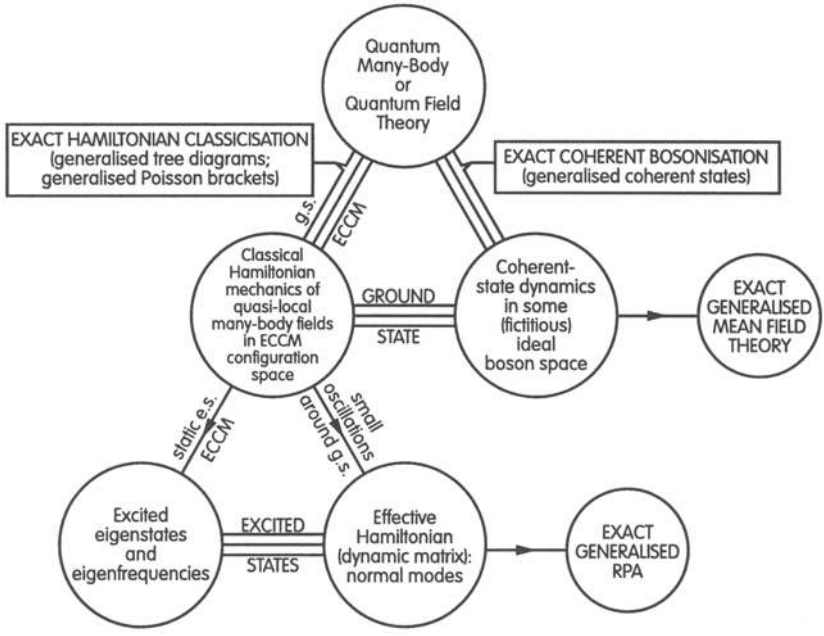

Fig. 2. A schematic representation of the hierarchical structure and the most general features of the ECCM.

we have attempted to describe above. In Figure 3 we have also attempted to make a very schematic overview of the hierarchical structure of the main elements of the CCM. We warn the reader that the schema shown is neither unique in its decomposition nor wholly rigorous or complete in the imposition of a logical structure on the resulting components. Nevertheless, we hope that Fig. 3 will serve, at least, as an *aide memoire* in surveying the formal aspects of what is, by now, a very wide-ranging formulation of microscopic quantum many-body theory.

We have already described, at least in outline, most of the CCM elements displayed in Fig. 3. We complete this Section with a very brief discussion of some additional items of coupled cluster theory, which largely reflect the work of the last few years or are the topics of current research.

CCM Parametrisation of Mixed States: Our entire discussion of the CCM up until this point has involved the parametrisation of quantum-mechanical pure states and, hence, is of relevance strictly to many-body systems at zero temperature. We note that an extension of the CCM to thermal mixed states, of relevance to systems at nonzero temperatures, was first given some ten years ago by Kümmel and his co-workers [133], in terms of the Bloch equation for the *statistical density operator*, $\hat{\rho} \equiv \mathrm{e}^{-\beta H} / \mathrm{Tr}(\mathrm{e}^{-\beta H})$,

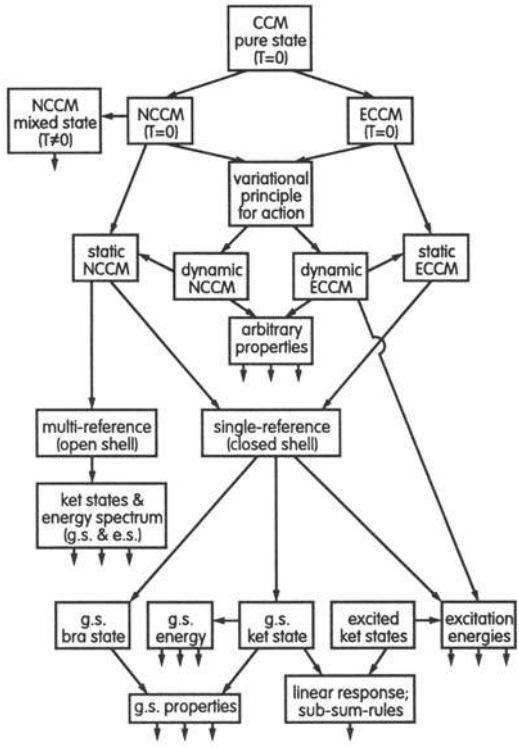

Fig. 3. A schematic diagram of the main ingredients of the CCM and their hierarchical structure.

where $\beta \equiv (k_{\mathrm{B}}T)^{-1}$ and T is the temperature. More recently, Mukherjee and his co-workers [134] have given a very natural and direct extension of the CCM to mixed states. In particular, they have provided a thermal cluster cumulant method for the grand partition function, $Z \equiv \mathrm{Tr}(\mathrm{e}^{-\beta(H-\mu\hat{N})})$, where μ is the chemical potential and $\hat{N}$ is the number operator. Unlike the earlier approach of Kümmel and his co-workers, the latter approach does not require a knowledge of the eigenspectrum of the many-body Hamiltonian H. For further details the reader is referred to the contribution by Mukherjee to the present volume.

Hermiticity-Preserving Versions of the CCM: As we have seen, both of the standard versions of coupled cluster theory (i.e., both NCCM and ECCM) are non-Hermitian by nature. The canonical coordinates, $\{s_I, \tilde{s}_I\}$ and $\{\sigma_I, \tilde{\sigma}_I\}$, which are intrinsically complex-valued for arbitrary wave functions, bear no simple relation to one another. In truncations to some subset of all available configurations $\{I\}$ the Hermiticity may be badly broken. Very

recently, Arponen [135] has complemented both versions of the CCM by introducing the complex conjugates of the cluster amplitudes. The resulting extended phase spaces are complex manifolds of too large a dimensionality. Arponen has shown how a set of *constraint functions* may be introduced so as to return to the *physical submanifold* (or reduced phase space, or constraint surface) which corresponds precisely to the original Hilbert or Fock space, $\mathcal{G}$. He thereby eliminates the extra degrees of freedom, by the Dirac bracket method, leaving as independent coordinates a minimal set of (eventually real) multiconfigurational amplitudes. These intrinsically Hermitian versions of the NCCM and ECCM hold out the promise of making practicable Hermiticity-preserving truncations available for practical applications.

General Fermionic CCM: Most many-body Hamiltonians H contain only products of an even number of fermionic configuration operators multiplied by ordinary (complex) numbers. The same is true for the multiconfigurational creation operators $\{C_I^\dagger\}$ considered up to this point. An example is provided by Sect. 2.3. However, for the purposes of calculating odd fermionic quantities such as Green's functions, correlation functions, or excited states, it is often convenient to break the fermionic parity of H, e.g., by the addition of *source terms* of the form $\sum_\alpha(\tilde{\mu}_\alpha a_\alpha + a_\alpha^\dagger \mu_\alpha)$. In such cases it is necessary to enlarge the commutation rules (14) to *graded commutation rules,* and to enlarge the corresponding set of CCM amplitudes, e.g., $\{s_I, \tilde{s}_I\}$ and $\{\sigma_I, \tilde{\sigma}_I\}$ from ordinary c-numbers to (complex) *Grassmann numbers* of both even and odd parity [119,136]. The procedure to do so is relatively straightforward, as has been demonstrated by Arponen [131].

Functional Form of the CCM: As we have seen above, the standard CCM parametrisations are operatorial in nature, where the ground-state wave functions, for example, are described in terms of correlation operators $(S, \tilde{S})$ or $(\Sigma, \tilde{\Sigma})$. However, as we have already mentioned in Sect. 1, similar functional forms of the CCM have also been employed recently [75–80,82], in which the ground-state ket wave function, for example, is specified in the exponential form, $\langle\{r\}|\Psi_0\rangle = \exp[S(\{r\})]$, where $|\{r\}\rangle$ is some suitably chosen complete set of wave functions. At corresponding levels of truncation the functional and operatorial versions are not identical, as has been discussed in detail in Ref. [82], to which the interested reader is referred for further details.

4 Applications of the CCM

Although our aim in the present article has been to review the general aspects of the coupled cluster method itself, we close with a very brief summary of some of the more important applications of the method that have so far been made. For further details of each application we refer the interested reader

to the references cited in each case, as well as to other reviews of the CCM [8,9,66–69,76,101,110,112,118,126,131,132,135,137–142].

Without any attempt to be exhaustive, we note that some of the most important applications of the CCM have been to the following systems:

- **Atomic Nuclei:** The NCCM has very successfully been applied to the ground and excited states of both closed-shell nuclei (e.g., ^{4}He, ^{16}O, ^{40}Ca) and to the open-shell nuclei (e.g., ^{15}N, ^{17}O, ^{14}C, ^{18}O, ^{18}F) formed from the addition to the closed-shell nuclei of one or two valence particles or holes. Calculations have been performed [8,9,70–75,100,107,122,143] for a variety of phenomenological two-body (and three-body) internucleon potentials, using the HCSUBn truncation scheme essentially up to the $n = 4$ level. Numerical convergence has been demonstrated at this level; and no other *ab initio* technique has ever bettered (or even equalled) these results, most of which are now about twenty years old.
- **Nuclear Matter:** Very similar calculations have also been performed [144] for nuclear matter, with similarly converged results for the binding energy per nucleon and the saturation density, for a range of phenomenological internucleonic forces.
- **Atoms and Molecules:** The CCM has become the method of choice in quantum chemistry wherever high accuracy is required. A huge range of calculations on such quantities as electron affinities, excitation energies, and ionisation potentials has been performed (see, e.g., Refs. [10,52,99, 105,106,123–125,137,139,140]) for a wide variety of atoms and molecules with up to about 100 active electrons. Especially for the heavier atoms, calculations have been performed with relativistic and QED corrections incorporated. The interested reader is referred also to recent reviews [145], and to the article by Kaldor in the present volume.[2]
- **The Electron Gas:** The NCCM has been applied to the electron gas in the high-density (RPA) limit, in the metallic density range, and in the low-density (Wigner crystal) regime. Results [112,146,147] in the important metallic density range for the correlation energy, for example, are accurate to better than 1 mH per electron (or $< 1\%$) by comparison to the essentially exact GFMC results [148] of Ceperley and Alder. This accuracy has never been equalled in any other *ab initio* calculation of what is still one of the best studied of all quantum many-body systems.
- **Charged Impurity in a Polarisable Medium:** The technique of allowing low-energy positrons to annihilate inside metals, alloys, and other forms of condensed matter has become an important experimental tool. Such systems comprising a charged impurity in a polarisable medium are prototypes of field-theoretical one-body problems. By casting the polarisation degrees of freedom as *internal gauge fields,* it has been shown [128]

[2] Issues 2–6 of Theoretica Chimica Acta **80** (1991) and a forthcoming issue of Molecular Physics (1998) are also wholly devoted to articles on coupled cluster theory and its applications.

how the ECCM can provide a powerful microscopic description of such systems.

- **Quantum Fluid Mechanics:** By coupling to *external gauge fields* (viz., the U(1) scalar and vector potentials), which serve to set the system into its most general state of motion, it has been shown [127] how the ECCM can provide a very complete, microscopic description of the zero-temperature quantum hydrodynamics of a strongly-interacting condensed Bose fluid.
- **Quantum Spin Lattice Models:** Extensive and very successful application of the CCM have been made in recent years to a wide variety of quantum spin lattice models. Examples include the solid phases of ^{3}He [149]; the isotropic (Heisenberg) and anisotropic XXZ models in one dimension and on the two-dimensional square lattice, both for spin-half systems [113–115,150] and higher-spin systems [116,151]; the spin-one Heisenberg-biquadratic model [152]; and to such *frustrated* spin models as the spin-half J_1-J_2 model [92,153], and the two-dimensional (anisotropic) Heisenberg antiferromagnet on a triangular lattice [117,118]. The CCM has also been applied to the spin-one easy-plane ferromagnet [154].
- **Electron Lattice Models:** The CCM has been applied [155] to such lattice models of strongly interacting electrons as the Hubbard model, both precisely at half-filling and with the further removal of one or two electrons.
- **Anharmonic Oscillators:** Both the NCCM and ECCM have been intensively applied to anharmonic oscillator systems [69,90,108,130–132,156] as $(0 + 1)$-dimensional models of nonlinear quantum field theories. The structure of the NCCM and ECCM has been particularly explored by applications to this model of the powerful holomorphic (or Bargmann) representation. Analogous anharmonic spin models have also been studied [90].
- **Continuum Quantum Field Theory:** Kümmel and his co-workers have very successfully applied the NCCM to several continuum quantum field theories. These include: (i) Φ^4 field theory [157] in $1 + 1$ (and $2 + 1$) dimensions, in the vacuum (ground state) and one-particle (mass gap) sectors, and in the soliton sector; (ii) $\Phi^4 + \Phi^6$ field theory [158] in $1 + 1$ dimensions, in the vacuum, one- and two-particle sectors; (iii) the sine-Gordon model in $1 + 1$ dimensions [159] in the vacuum, one-particle, and soliton sectors; and (iv) the vacuum, one-particle (physical nucleon mass), and two-particle (deuteron) sectors of a model $(3 + 1)$-dimensional field theory of pions and nucleons [160] interacting via the usual isospin-invariant pseudoscalar coupling. We also note that a preliminary application of the ECCM to quantum field theory has also been made [161], using the sine-Gordon model as an example.
- **Lattice Quantum Field Theory:** As discussed in Sect. 1, the NCCM has been widely applied to various lattice gauge field theories. These

include the discrete Z(2) model [75], and such continuous groups as the Abelian U(1) case [75-80] and the non-Abelian SU(2) case [81]. Both the operatorial and functional forms of the NCCM have also been applied very recently [82] to a latticised O(4) nonlinear sigma model of chiral meson field theory.

- **Quantum Optics:** A very recent application of the NCCM has been made to one of the prototypical models of quantum optics, namely the Rabi Hamiltonian (or Jaynes–Cummings model without the rotating wave approximation) [162].
- **Solid-State Optoelectronics:** A similar application of the NCCM has been made to the linear *E-e* Jahn–Teller effect [163].
- **Other Model Problems:** Various other applications of the CCM have been made to such important model problems as the Lipkin–Meshkov–Glick model referred to [89] in Sect. 2.4 [67,107,129,164]; and the one-dimensional Lieb model [86,87] of bosons interacting via repulsive delta-function potentials [84].

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