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Origins of the Coupled Cluster Method

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Summary. The origins of the coupled cluster method are traced back to the necessity of dealing with the hard core potentials of nuclear physics. The exponential structure of the wave function follows from very general arguments: using the Bargmann representation it appears as solution of a simple differential equation. Or, using plausibility arguments, the virtual excitations out of the Fermi sea naturally show up as exponentials. The unresolved convergence problems are discussed. I believe that the title of this talk gives me some freedom in the selection of the topics I may speak about. Thus I chose to say only very little about the history and more about the physical background which with necessity lead to the coupled cluster (CC) method. Just this necessity will be a central point of my talk. Indeed I shall make some remarks, ranging from plausibility arguments to rigorous proofs, concerning the structure of ground state wave functions. I shall say very little about open shell systems. Here the CC techniques have been developed much later, are less unique and my own contributions are relatively few.

Key words: Coupled Cluster Method – Perturbation theory – Bargmann space – Convergence

1. Perturbation theory and Gell-Mann Low equation

It all started in the mid fifties when people did believe that it should be possible and that it was highly time to solve the nuclear many body problem. "Possible" since computers at that time became a standard outfit of universities. Furthermore, at that time two body forces became available, derived from two nucleon scattering experiments. Also one had a rather successful shell model which seemed to overcome the first difficulty that – contrary to the situation in atoms and molecules – there is no defining central field on which to build an independent particle description. Experimental phase analysis allowed (and even today still allows!) for many different internucleon potentials. Nevertheless all of them have a long ranged attractive tail (due to the pion exchange, leading to a range of the order of the Compton wave length of the pion, that is about 1.5 Fermi) and a short ranged repulsive core (due to exchange of heavier mesons, with a range of the order of 0.5 Fermi), which may even be infinite ("hard core" potential). This practically rules out a Hartree–Fock approximation since the potential matrix elements become very large or infinite. This was the second problem encountered. Using nuclear matter as a model (to have simple plane waves as s.p. basis) this problem was solved by Brueckner [1]: he summed up all "upgoing" ladder diagrams as shown in Fig. 1.

This is equivalent to solving by iteration the equation:

$$G = V + V\frac{Q}{e}G \tag{1}$$

for the "G-matrix", obtaining:

$$G = V + V\frac{Q}{e}V + V\frac{Q}{e}V\frac{Q}{e}V + \cdots. \tag{2}$$

Here Q is a projection operator excluding intermediate downgoing (hole) lines and e is an energy denominator. The correlation energy due to this ladder sum is $\Delta E \approx \frac{1}{2}\sum \langle v_1 v_2|G|v_1 v_2\rangle$, where $v_{1,2}$ label occupied states. In a next step Goldstone [2] rewrote this as a ("Bethe–Goldstone") equation for an amplitude χ_2:

$$(T_1 + T_2 - \epsilon_{v_1} - \epsilon_{v_2})|\chi_2|v_1 v_2\rangle = -QV|\chi_2|v_1 v_2\rangle. \tag{3}$$

Here $\epsilon_{v_{1,2}}$ are single particle energies for occupied states. The G-matrix is obtained simply as

$$\langle \alpha\beta|G|v_1 v_2\rangle = \langle \alpha\beta|V\chi_2|v_1 v_2\rangle. \tag{4}$$

From this it is rather evident that summing up the ladder amounts to solving a (Schrödinger type) *differential equation with the boundary condition that the function must vanish inside the hard core* (if any). This boundary condition is well known to have a solution. There is only the technical problem of dealing with the operator Q, which nowadays is routine. Even in the most sophisticated calculations for hard core or similar problems this approximation typically serves as a starting point. Brueckner already did improve on it by incorporating some self energy terms: he did replace the single particle energies by

$$\epsilon_{v_i} \to \epsilon_{v_i} - \sum_{v} \langle v_i v|G|v_i v\rangle \equiv \epsilon_{v_i} - \sum_{v} \langle v_i v|V\chi_2|v_i v\rangle.$$

Thereby the Bethe–Goldstone equation did become a nonlinear equation to be solved self consistently. Clearly the hole lines have been "dressed" by adding this term, see Fig. 2.

The experts know that this is just a subset of the terms in the two body equation of the CC method. I have written this equation in pictorial form in Fig. 3. (The dash-dotted line indicates the energy denominator.) Here

$$\chi_2|v_1 v_2\rangle = |v_1 v_2\rangle + S_2|v_1 v_2\rangle$$

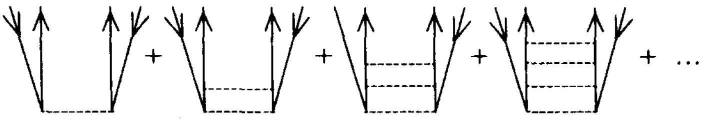

Fig. 1. Brueckner ladders

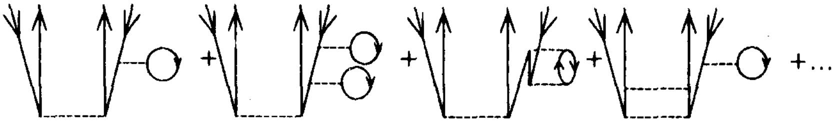

Fig. 2. Brueckner ladders with hole line insertions

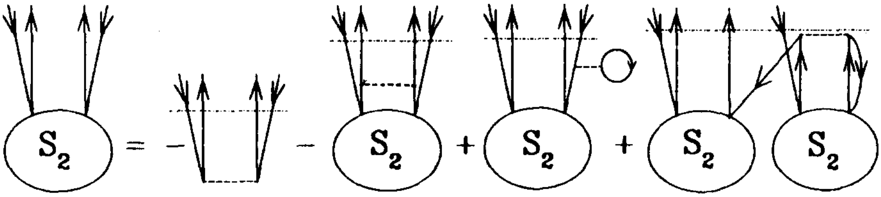

Fig. 3. S_2 equation generating Brueckner ladders

is used. The first part is the determinant of two single particle states, the second one is the well-known S_2 amplitude. Inside the hard core the boundary condition:

$$\langle x_1 x_2 | S_2 | \nu_1 \nu_2 \rangle = -\langle x_1 x_2 | \nu_1 \nu_2 \rangle$$

has to be observed. The first two terms on the right hand side of Fig. 3 generate the ladders and the last one dresses the hole lines.

At the same time Goldstone did prove the linked cluster theorem by carefully inspecting the perturbation series named after him, although Brueckner had found before that the lowest order unlinked terms cancel. As seen from today one should be aware of the fact that the linked cluster theorem is "trivial" because (for extended systems) the energy contributions individually are proportional to the particle number N and thus products of n such terms go as N^n. Shortly afterwards Hubbard [3] and a little later Hugenholtz [4] did establish the exponential structure, again by skilful analysis of perturbation theory. I shall not go into any technical details, mainly because I never managed to arrive at a simple insight using this approach. Indeed, from Fig. 3 nobody will recognize *a priori* that it is generated by the usual manipulation with $\exp(-S)H\exp S$. Only if one *starts* with the exponential one may derive those terms *a posteriori.*

At this place I want to draw your attention to a much earlier paper by Gell-Mann and Low [5] which entered into some textbooks on quantum field theory, e.g. [6]. As far as I know it is indeed the first one to arrive at an exponential form. This is based on the standard time dependent perturbation theory of quantum field theories, using the interaction representation. A bound state wave function and energy turns out to be:

$$|\Psi_0\rangle = \frac{U(0,-\infty)|\Phi_0\rangle}{\langle\Phi_0|U(0,-\infty)|\Phi_0\rangle} \quad \text{and} \quad E = \frac{\langle\Phi_0|HU(0,-\infty)|\Phi_0\rangle}{\langle\Phi_0|U(0,-\infty)|\Phi_0\rangle}. \tag{5}$$

Here $U(0,-\infty)$ is the time development operator in the interaction representation for the time $-\infty$ to 0 and Φ_0 the bare vacuum in field theory or the Slater determinant in a many body system. Now it is well known that U

can be expanded as:

$$U(0, -\infty) = \sum_{0}^{\infty} \frac{(-i)^n}{n!} \int_{-\infty}^{0} \cdots \int_{-\infty}^{0} T[V(t_1)V(t_2) \ldots V(t_n)]\, dt_1\, dt_2 \ldots dt_n$$
$$= T \exp\left(\int_{-\infty}^{0} dt\, V(t)\right). \tag{6}$$

T is the usual time ordering operator. The perturbation series (6) represents modified Feynman diagrams, the non-covariant versions of which are the Goldstone diagrams. In field theory it is well known that the terms are linked: The denominator $\langle \Phi_0 | U(0, -\infty) | \Phi_0 \rangle$ just cancels the unlinked vacuum-vacuum diagrams occurring in the numerator. I mention in passing that there is no proof that this wave function really exists, although there should be little doubt for finite many body systems. There is still some way to go to the exponential form as used in the CC method, but clearly it is buried in this expression.

Returning now to history, the next big step was a short paper by Fritz Coester [7]. Knowing Hubbard's paper he used the exponential form as a vehicle to derive the linked cluster structure, had the idea to compute *amplitudes* directly instead of perturbation terms and he obtained formal equations for them. This was the time I myself appeared at the scene. If I remember correctly we then realized that the Bethe–Goldstone and the corresponding three body ("Bethe–Fadejev") equations were embedded in this set of equations. Years later Lührmann and myself [8] managed to derive rigorously the n-body equation and got the insights needed for dealing with hard core potentials in arbitrary order. From then on it was mainly technical work, where John Zabolitzky was the most prominent figure. Ray Bishop will talk about the applications [9] and thus I may stop here with history.

Except for one curious fact: for about ten years nobody in the nuclear physics community did care for the CC method. People were partially right since the computers at that time were just too small to make use of more than the lowest order approximations. Thus nobody did care for the strength – and the beauty – of the method, namely the systematics behind it and the unique truncation schemes it therefore suggests. Indeed for hard core potentials there is practically no freedom in truncating the set of CC equations! It was thus a happy surprise to me as I opened a big parcel with reprints sent to me by Jiri Cizek in the mid-1970s telling me that the chemist not only had read our papers, they even were daring enough to try some applications. Indeed Cizek was the first one to make explicit the approximate CCM equations and to use them for chemistry problems [11]. The first *ab initio* calculations were done by Paldus et al. [12]. Since this is not the topic of my talk, I don't go into any more details. Instead, I refer to Bartlett's review [13] and papers presented at this workshop. As much as the chemists must be praised for occasionally looking into nuclear physics journals, as much I have to regret that nuclear physicists (including myself) did not care to search in the other direction. This hasn't changed much, especially the particle physicists still believe that the higher the energy they are dealing with the lesser they need to look down to the low energy people.

2. Bargmann space and exponential form

Now let me illustrate why history had to take this course, that is why one had to arrive at the CC method. Strange as it may be, in spite of the many successes of the coupled cluster method there is still a widespread belief that the underlying exponential structure is something artificial, accidental or even an approximation only. This is why I want to make clear that this feature is extremely natural – even necessary – on a very fundamental level, not necessarily connected with many body theory.

I am using first some old arguments put forward by Fritz Coester and Rudolf Haag as far back as 1960 [10]. I modify and extend this a bit since it was originally formulated for (bosonic) quantum field theories and at that time the Bargmann space was not known to the physics community. For the moment I restrict myself to bosons. Assume that on the one hand everything may be represented by the Fock space with the usual creation and annihilation operators $\boldsymbol{a}_i^\dagger$ and $\boldsymbol{a}_j$ and a bare vacuum $|\Phi_0\rangle$. Actually this is not necessary but helps to relate the following points to more familiar things. On the other hand, in the Bargmann representation these operators are replaced by complex numbers as follows:

$$\boldsymbol{a}_i^\dagger \rightarrow z_i \quad \text{and} \quad \boldsymbol{a}_j \rightarrow \frac{\partial}{\partial z_j}. \tag{7}$$

The analytic functions of $z_1 z_2 \ldots f(z_1 z_2 \ldots)$ (abbreviated as $f(z)$) generate a Hilbert space with the scalar product:

$$\langle f|g\rangle \equiv \prod_i \left(\int \frac{dz_i\, d\bar{z}_i}{2\pi i} \exp(-z_i\bar{z}_i)\right)\bar{f}(\bar{z})g(z). \tag{8}$$

The functions:

$$|n_1 n_2 \ldots\rangle = \prod_i \frac{(\boldsymbol{a}_i^\dagger)^{n_i}}{\sqrt{n_i!}}|\Phi_0\rangle \rightarrow \prod_i \frac{z_i^{n_i}}{\sqrt{n_i!}} \tag{9}$$

define an orthonormal basis in this space. Clearly they are counterparts of Fock space states with n_i particles in the state labelled by i. Making the assumption that the given Hamiltonian $\boldsymbol{H}$ has a (discrete normalizable) ground state Ψ_0, it can be shown that an arbitrary state can be written as

$$\Psi(z) = G(z)\Psi_0(z). \tag{10}$$

This equation is trivial, since for bosons the ground state has no zeros.

But then also the state created by $\boldsymbol{a}_i$ must have this feature:

$$\boldsymbol{a}_i\Psi_0 \rightarrow \frac{\partial}{\partial z_i}\Psi_0(z) = L_i(z)\Psi_0(z), \tag{11}$$

the solution of which is:

$$\Psi_0(z) = \exp S(z) \quad \text{with} \quad L_i(z) = \frac{\partial}{\partial z_i}S(z). \tag{12}$$

Here a general $S(z)$ is a superposition of polynomials:

$$S(z) = \sum_n S_n(z) \tag{13}$$

with

$$\begin{aligned} S_n(z) &= \sum_{i_1, i_2, \ldots, i_n} \frac{1}{n!} S_n(i_1, i_2, \ldots) z_{i_1} z_{i_2} \ldots z_{i_n} \\ &= \frac{1}{n!}\int \cdots \int dx_1 \ldots dx_n\, S_n(x_1 \ldots x_n) z(x_1) \ldots z(x_n), \end{aligned} \tag{14}$$

where in the last step we did switch to the coordinate representation. Clearly this is the Bargmann space analogue of the exponential form as used in the coupled cluster method. This result is quite general, and valid even for a one body problem. It is remarkable but not at all surprising that the exponential form was so powerful even for the anharmonic oscillator [14].

We now turn to extended many body systems. Assume that the Hamiltonian is of the usual form with interactions of finite range. Then we may divide the space into (large) cells. To each cell belong operators $\boldsymbol{a}^{(k)\dagger}(x)$ and $\boldsymbol{a}^{(k)}(x)$ (variables $z^{(k)}(x)$) restricted to the cell k. The Hamiltonian becomes

$$\boldsymbol{H} \approx \sum_k H_k, \tag{15}$$

and the wave function Ψ_0 becomes a product

$$\Psi_0(z) \approx \prod_k \Psi_0(z^{(k)}), \tag{16}$$

where

$$z^{(k)}(x) = z(x) \quad \text{if } x \text{ in the cell } k, \quad = 0 \text{ otherwise}. \tag{17}$$

Hence

$$S(z) = \sum_k S(z^{(k)}). \tag{18}$$

Clearly the exponential form was essential for this "*cluster property*" of the wave function. Then Eq. (18) implies that $S_n(x_1 \ldots x_n)$ tends to zero with increasing distance between any points x_i. This connection between cluster structure and exponentials has a long history in statistical mechanics. The well-known Ursell–Mayer linked cluster expansion of the partition function uses the same ideas about the asymptotic behaviour of multidimensional fields.

Let me now turn to fermions. If one so wishes one may combine Bargmann space with Grassmann variables to take care of the anticommutating operators. But this would require going deeper into the underlying mathematics. Thus I shall present a more modest approach. Let Φ_0 be the (Slater) determinant of occupied states and $A_i^\dagger$ and A_i the creation operators with respect to a suitably chosen Φ_0 as "vacuum". Then without loss of generality the ground state wave function can be written as:

$$\Psi_0(A^\dagger) = F(A^\dagger)\Phi_0. \tag{19}$$

Here $F(A^\dagger)$ indicates a functional of the creation operators $A_i^\dagger$ with even number of such operators (with numbers of holes equal to number of particles because of particle number conservation). We show first that an arbitrary state can be written as:

$$\Psi(A^\dagger) = G(A^\dagger)\Psi_0. \tag{20}$$

One has to convince oneself that in Fock space one may generate an arbitrary vector out of Ψ_0 by creation operators alone. This always is possible since one easily may remove any undesirable particle by applying the corresponding creation operator, using $\boldsymbol{a}_i^{\dagger 2} = 0$. But then also:

$$A_i\Psi_0 = L_i(A^\dagger)\Psi_0 = [A_i, F(A^\dagger)]\Phi_0 = \frac{\delta F(A^\dagger)}{\delta A_i^\dagger}\Phi_0 = L_i(A^\dagger)F(A^\dagger)\Phi_0, \tag{21}$$

with the solution:

$$F(A^\dagger) = \exp[S(A^\dagger)], \qquad L_i(A^\dagger) = \frac{\delta S(A^\dagger)}{\delta A_i^\dagger}, \tag{22}$$

as desired. With suitable modifications all further conclusions can be taken over from the bosonic case.

3. Fermi sea and exponential structure

Let us now turn to simple plausibility arguments leading again to the exponential form, arguments which have been put forward before [15]. We want to determine the ground state wave function Ψ_0 and energy E of a system of N fermions, say, for the lowest eigenvalue E of the N-fermion system. For ground states of closed shell systems an important part of the wave function is the Slater determinant of occupied states $|\Phi_0\rangle$. The underlying picture here is of course that each particle moves independently in a potential well produced by (the nucleus in the case of atoms and) the averaged motion of the other particles, which themselves otherwise move independently except for the restriction of quantum statistics imposed in this case by the Pauli principle. Due to the Pauli principle each electron is in a different single-particle eigenstate of this mean potential and the N lowest single particle states – representing the "Fermi sea" – are filled from below as indicated in Fig. 4a.

This Slater determinant of the N lowest single particle wave functions (the occupied states) may for instance be determined by the Hartree–Fock variational principle. But of course this picture of dynamically independent motion is not complete, since the particles will in general interact among themselves and hence cannot move independently. We now attempt to correct for these dynamic correlations systematically. The first thing one may imagine happening is that two particles mutually interact, thereby lifting themselves out of the Fermi sea, so that after the interaction both are in unoccupied orbitals, see Fig. 4b. This process will be described by some quantum-mechanical amplitude or equivalently by an operator S_2 which acts on the Fermi sea wave function $|\Phi_0\rangle$ to produce the wave function $S_2|\Phi_0\rangle$ describing two particles outside the Fermi sea (and consequently two "holes" inside it) and all remaining $N-2$ particles in their previous orbitals. But, it may also occur for example that two pairs of particles do this *completely independently*, as illustrated in Fig. 4c. This process is clearly 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 counting pairs twice. The resulting contribution to the wave function is thus $\frac{1}{2}S_2^2|\Phi_0\rangle$. This process of independent pair excitation out of the Fermi sea may be continued to obtain a contribution $(1/m!)S_2^m|\Phi_0\rangle$ for the amplitude describing the excitation of m independent pairs. Using the superposition principle we get

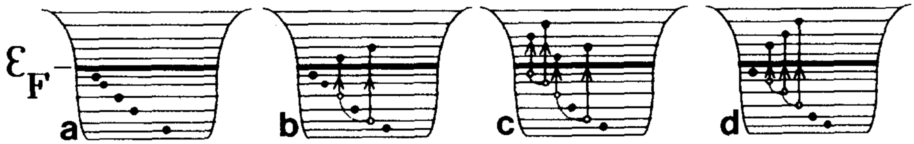

Fig. 4. Excitations out of the Fermi sea

the total amplitude

$$\sum_{m=0}^{\infty} \frac{1}{m!} S_2^m |\Phi_0\rangle = \exp(S_2)|\Phi_0\rangle. \tag{23}$$

Continuing in this way, we next imagine the simultaneous excitation of three particles, as in Fig. 4d, which will be described by a contribution $S_3|\Phi_0\rangle$. Similarly there will be a contribution $(1/n!)S_3^n|\Phi_0\rangle$ from the simultaneous excitation of n independent triplets. Next, we can also imagine the simultaneous excitation of m pairs and n triplets all independently from each other, with an amplitude $(1/n!m!)S_2^m S_3^n|\Phi_0\rangle$. Note that the operators S_2 and S_3 commute since they describe independent processes. Summing over all n and m we arrive at $\exp(S_2+S_3)|\Phi_0\rangle$ as contribution from all pair and triplet excitations. Proceeding in this way with the excitation of clusters of 4, 5, . . . , N particles we arrive at a wave function $\exp(S_2+S_3+\cdots+S_N)|\Phi_0\rangle$. But it also may happen that during the interaction of any subset of particles only one of them is finally lifted out of the Fermi sea. Also any number of single particles may independently be promoted out of the Fermi sea. As before we are led to describe this process by an operator $\exp(S_1)|\Phi_0\rangle$, where S_1 acts on $|\Phi_0\rangle$ to produce a single "particle-hole pair" (or excitation) on the Fermi sea. This particular case has a special meaning provided by the Thouless theorem [16]. It states that the most general determinantal wave function $|\Phi_0'\rangle$ not orthogonal to a given Slater determinant $|\Phi_0\rangle$ has the form $\exp(S_1)|\Phi_0\rangle$. In other words the effect of the independent elevation of single particles is equivalent to changing the single particle orbitals.

We thus arrive at the wave function $|\Psi\rangle = \exp(\sum_{n=1}^{N} S_n)|\Phi_0\rangle$, together with a physical interpretation of both the exponential form and the individual operators S_n.

4. Final remarks

I have stressed very much the exponential structure. But equally important is of course the idea to directly compute the amplitudes in it. We all know that this cannot be done exactly. Approximations have to be made. One of the strength's of the CC method is that it leads to fairly unique truncation schemes, certainly for hard core potentials and to a lesser degree also for smooth potentials. There is one important aspect – for fermions often supported by numerical evidence – that a sum of terms growing out from one term through antisymmetrization should be kept together, i.e. taken along or thrown away together. The sum of all those terms often is much smaller than each individual term. One cannot always live up to this ideal of taking along them all, because certain multidimensional integrations or corresponding summations cannot be performed. A judicious choice of the terms neglected typically will suffice, since in a systematic approximation the "questionable" terms will anyway be small. To make these remarks less abstract let me refer to our nuclear physics calculations: for evident reasons we could not include all four body terms. But those we could take along individually were of the order of 1% of the total energy, and their sum – due to the antisymmetrization – was of the order of only 0.2%. The remaining untreatable terms could be estimated to be again of the order of 1%. The net result was that the four body terms were 1% corrections, but we could not reliably calculate them. For the chemists it should be said that for nuclei this is an extremely high accuracy.

The least understood aspect of the CC method is the question of convergence. All indications are that one has at most asymptotic convergence. Jouko Arponen [17] will say something about it. I myself recall some lively discussions I had with Fritz Coester on this topic. One of them ended with his remark: "I don't understand why the CC method is successful. *It should not work*".

The problem is that rigorous mathematical analysis at present is possible only in one dimensional systems. Conclusions drawn from this taken over to many degrees of freedom very likely are wrong. I guess that they are too pessimistic. Especially the antisymmetrization for fermions reduces the total size of sets of terms as mentioned before. I have no idea how one could get a firm mathematical grip on this phenomenon. But the same applies to all perturbative methods I know about. Of course, I believe that numerical evidence for convergence justifies the application of the method.

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