Near-degenerate systems in nuclear structure and quantum chemistry from ab initio many-body methods

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I. PROBLEM

The present interdisciplinary program builds on the fact that the ab initio description of near-degenerate finite fermi systems with an open-shell character currently constitutes a frontier in both low-energy nuclear theory and quantum chemistry. It is thus believed that both theory communities can strongly benefit from cross-disciplinary activities and the present workshops aims at acting as a jump-start to move in this direction.

Ab-initio many-body methods are nowadays capable of tackling nuclei up to masses $A \sim 130$ and molecules up to a few hundred electrons. As for nuclei, this limit has been achieved only recently in converged calculations with realistic two- and three-nucleon interactions. Pivotal to reaching medium mass nuclei was the introduction of techniques such as coupled cluster (CC) theory [1–3], self-consistent Green's function (SCGF) theory [4–6], and in-medium Similarity Renormalization Group (IMSRG) theory [7, 8]. A major achievement has been the extension of SCGF [9] and CC [10, 11] formalisms to tackle realistic three-nucleon forces, which allows now the accurate testing of nuclear interactions. However, these methods have until recently been limited to closed-shells systems and those accessible by the addition/removal of 1 or 2 particles (electrons or nucleons). This, de facto, limits their applicability for studies of excitation spectra and excludes the vast majority of isotopes of current interest at nuclear RIB facilities.

The extension to truly open-shell nuclei constitutes a major hurdle to overcome as it requires expanding of the many-body problem around a degenerate reference state (i.e. the uncorrelated state upon which the many-body method is constructed, usually a Hartree-Fock wave function). Still, it is a key challenge that must and is being

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undertaken in view of the wealth of data expected from new radioactive beam facilities, especially regarding neutron-rich nuclei. Extending any of the available many-body methods to open-shell systems generates an extra layer of complexity in the formalism and a scaling up in the computational cost. There are typically two ways to attack the problem, i.e. (i) extend the reach of the method through a multi-reference formulation or (ii) reformulate the expansion around a single symmetry-broken reference state to efficiently capture the dominant part of collective correlations (typically, particle-number breaking tackles pairing in singly open-shell nuclei while rotational symmetry must be further broken to embrace quadrupole correlations in doubly open-shell nuclei). The first path maintains good symmetries throughout but is complicated by the need to mix different reference states. The latter path requires a non-trivial step to eventually restore the symmetries that are broken in the first place.

In the nuclear context, a MR method has been recently proposed within the IMSRG framework to tackle (singly so far) open-shell systems [12]. Even more recently, CC-based [13] and IMSRG-based [14] ab-initio configuration interaction methods have been proposed. The SCGF method also has the potentiality to yield configuration interaction of the same quality along with consistent effective charges [15]. These advances provide a direct path to eventually link realistic forces (that are hopefully consistent with the underlying QCD) to nuclear properties and electroweak responses of nuclei that are or will be under investigation at RIB facilities. Nevertheless ab-initio MR methods for nuclei are still in their infancy and can benefit from a confrontation with the more advanced quantum chemistry formulations (see below).

The path based on the spontaneous breaking of symmetries is currently being followed for singly open-shell nuclei through the Gorkov extension [16, 17] of Green's function theory and the recently formulated Bogoliubov coupled cluster theory [18, 19]. These ab-initio calculations of singly open-shell nuclei are now a reality and have addressed long isotopic chains around, e.g., Ca and Ni. However, these approaches are not yet mature and they need to be developed further to reach the same accuracy as their closed-shell counterparts and to tackle doubly open-shell systems, which is the next major challenge for ab-initio nuclear theory. Additionally, while the breaking of symmetries is an efficient way to bypass near-degeneracy problems by expanding the many-body solution around a single product state (i.e. a symmetry unrestricted Slater determinant or its Bogoliubov extension) with a closed-shell character, symmetries must be eventually exactly restored when dealing with finite quantum systems. This is mandatory to have the correct energetics, interpret the spectroscopy meaningfully and compute amplitudes of, e.g. electromagnetic, operators between the associated states. At the strict mean-field level, symmetry restoration techniques have been heavily developed in nuclear physics over the last twenty years [20] and recently imported in quantum chemistry to tackle static correlations efficiently [21]. Very recently, the exact restoration of broken symmetries beyond the (projected) mean-field level has been formulated for the first time within the frame of ab initio CC theory [22]. It is of interest to investigate whether such an approach can be extended to self-consistent (Gorkov) Green's function theory.

Coupled-cluster theory [23] is the gold standard of quantum chemistry and is now well developed for nearly any single-reference state. However, even if most ground states of molecules are of closed-shell nature, they acquire an open-shell character as they undergo bond breaking. To bypass difficulties associated with near-degenerate systems, the benefit of using a symmetry-broken reference has long been recognized in quantum chemistry in general and for coupled cluster calculations in particular. Still, high-order coupled cluster results often remain contaminated by the breaking of the symmetry [24], e.g. spin characteristics are not preserved, which compromises the energetics and the predictions of magnetic properties, as well as the interpretation of excited states as already alluded to above. Part of the solution is given by using equation-of-motion [25, 26] (EOM) methods, to add or remove electrons from a nearby closed-shell reference state with correct spin. This automatically generates proper spin eigenfunctions. The same benefit is obtained in the nuclear context [27]. For genuinely open-shell molecules it is however usually impossible to find an appropriate reference state close enough. This calls for an exact restoration of the broken symmetry, which could be achieved on the basis of the development recently made in the nuclear context [22].

When the single reference approach fails, another route toward enabling coupled cluster methods to describe open-shells correctly, while preserving the symmetries throughout, is to develop a multi-reference (MR-CC) approach that treats several equally important determinants as a "static correlation" reference [28, 29]. As a molecule dissociates, the optimal reference state can change drastically confirming the necessity of a MR description. As bond-breaking and forming is the very essence of chemistry, the importance of a MR description cannot be overestimated. As a matter of fact, several problems in addition to bond forming and breaking, i.e. transition metal multiplets and excited states cannot be described as well as required without a MR description. Furthermore, applications to strongly correlated systems are beyond the capability of the single reference coupled cluster theory. Consequently, the development of MR methods constitutes a very active area of research with several different routes currently being pursued, but no obvious winner has as yet emerged, knowing that the different approaches have to also contend with issues beyond symmetry.

II. GOALS OF THE WORKSHOP

In summary, the goals of the workshop are to

1. bring together nuclear theorists and quantum chemists facing similar challenges regarding the description of open-shell systems,
2. present state-of-the-art methods based on multi-reference or symmetry-breaking-and-restoration schemes,
3. identify the optimal MR methods in terms of theory and application for various problems of interest in quantum chemistry and nuclear physics,
4. understand in what situation MR approaches are preferable to symmetry-breaking-and-restoration schemes and vice versa.

In order to facilitate the understanding and involvement of young theorists and experimentalists as well as to lay a common ground for quantum chemists and nuclear theorists, four 1h15mns lectures on the basics of (i) symmetry broken and restored mean-field theory, (ii) coupled-cluster theory, (iii) self-consistent Green's function theory and (iv) in-medium similarity renormalization group theory, will be given at the beginning of the workshop.

III. PROGRAM

A. Introductory lectures

1. C. A. Jiménez-Hoyos, *Symmetry broken and restored mean-field theory*
2. S. Bogner, *Basics of single-reference and multi-reference in-medium similarity renormalization group theory*
3. V. Ortiz, *Basics of self-consistent Green's function theory*
4. R. J. Bartlett, *Basics of single-reference and multi-reference coupled cluster theory*

B. Workshop speakers

1. Multi-reference many-body theories
   - H. Hergert, *Multi-reference in-medium similarity renormalization group theory for nuclei*
   - A. Koehn, *Internally contracted multi-reference coupled-cluster method*
   - M. Musial, *Fock space multi-reference coupled-cluster method*
2. Effective operators for ab-initio configuration interaction calculations
   - J. D. Holt, *Non-perturbative shell-model interactions from the in-medium similarity renormalization group*
   - G. Hagen, *Ab-initio coupled-cluster effective interactions for the shell model*
   - C. Barbieri, *Effective operators for shell-model calculations from self-consistent Green's function theory*
3. Symmetry-unrestricted many-body theories
   - V. Somà, *Self-consistent Gorkov Green's function theory for nuclei*
   - A. Rios, *Self-consistent Green's function with anomalous propagators for homogeneous nuclear matter*
   - T. M. Henderson, *Bogoliubov coupled cluster theory for the attractive pairing Hamiltonian*
   - A. Signoracci, *Bogoliubov coupled cluster theory for nuclei*
4. Symmetry- (broken and) restored many-body theories
   - M. Bender, *Symmetry-restored mean-field theory for nuclei*
   - T. Duguet, *Symmetry-restored coupled cluster and self-consistent Green's function theories*

5. Reaching excited states and/or neighboring systems
   - P. Piecuch, *Molecular systems from the equation-of-motion coupled-cluster theory*
   - G. R. Jansen, *Near closed-shells nuclei from the equation-of-motion coupled-cluster theory*
6. Additional flavors
   - F. Evangelista, *The similarity renormalization group in quantum chemistry*
   - J. Toulouse, *QMC trial wave functions and their optimization for chemistry*
   - G. E Scuseria, *Low-cost generalized coupled cluster models for strong and weak correlations*
   - L. Reining, *A direct approach to the calculation of many-body Green' s functions*

C. Discussion sessions

1. Discussion 1: *Multi-reference methods*
2. Discussion 2: *Symmetry broken and restored methods*
3. Discussion 3: *Possible cross-disciplinary projects between quantum chemistry and nuclear physics*

D. Schedule

Lectures	**Workshop**		
Mon. Mar. 30th	*Tue. Mar. 31st*	*Wed. Apr. 1st*	*Thu. Apr. 2nd*
Room 135	*Room 135*	*Room 135*	*Room 135*
09h15 *Welcome*	09h30 Hergert	09h30 Signoracci	09h30 Toulouse
09h30 Jimenez-Hoyos	10h15 **Break**	10h15 **Break**	10h15 **Break**
10h45 **Break**	10h45 Koehn	10h45 Henderson	10h45 Bender
11h15 Bogner	11h30 Evangelista	11h30 Scuseria	11h30 Duguet
12h30 **Lunch**	12h15 **Lunch**	12h15 **Lunch**	12h15 **Lunch**
14h15 Ortiz	14h00 Barbieri	14h00 Hagen	14h00 Piecuch
15h30 **Break**	14h45 Holt	14h45 Soma	14h45 Jansen
16h00 Bartlett	15h30 **Break**	15h30 **Break**	15h30 **Break**
17h15 **End**	16h00 Musial	16h00 Reining	16h00 Rios
	16h45 Discussion 1	16h45 Discussion 2	16h45 Discussion 3
	18h00 **End**	18h00 **End**	18h00 **End**
	20h00 **Dinner**		

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