Coupled-Cluster Approach to Electron Correlations in the Two-Dimensional Hubbard Model

Yoshihiro Asai and Hideki Katagiri
Electrotechnical Laboratory (ETL),
Agency of Industrial Science and Technology (AIST),
Umezono 1-1-4, Tsukuba, Ibaraki 305, Japan
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We have studied electron correlations in the doped two-dimensional (2D) Hubbard model by using the coupled-cluster method (CCM) to investigate whether or not the method can be applied to correct the independent particle approximations actually used in ab-initio band calculations. The double excitation version of the CCM, implemented using the approximate coupled pair (ACP) method, account for most of the correlation energies of the 2D Hubbard model in the weak ($U/t \simeq 1$) and the intermediate $U/t$ regions ($U/t \simeq 4$). The error is always less than 1% there. The ACP approximation gets less accurate for large $U/t$ ($U/t \simeq 8$) and/or near half-filling. Further incorporation of electron correlation effects is necessary in this region. The accuracy does not depend on the system size and the gap between the lowest unoccupied level and the highest occupied level due to the finite size effect. Hence, the CCM may be favorably applied to ab-initio band calculations on metals as well as semiconductors and insulators.

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Electron correlation in solids is the origin of various macroscopic quantum phenomena such as magnetism and high-$T_c$ superconductivity. Interests in these fields are gradually shifting from materials which have simple chemical compositions and structures to more complicated ones. Besides the importance of simplified models such as the Hubbard and Heisenberg models in statistical physics, the importance of approximate many-body theories which can be applied to ab-initio calculations is rapidly increasing.

The coupled cluster method (CCM) has been recognized as one of the most successful approximate methods for quantum many-body problems in nuclear physics and ab-initio quantum chemistry. The method has also been successfully applied to the electron gas problem, but there have been few applications to quantum lattice problems. Within the limited number of examples in the literature, we find that the method was successfully applied to quantum spin models, the one-dimensional Hubbard and Pariser-Parr-Pople (PPP) model, and the two-dimensional (2D) Hubbard model at half-filling and with one hole. These successful results suggest the CCM may be one of the most suitable methods to study electron correlations in solids as well as molecules and nuclei.

To verify this expectation, we have made more rigorous tests of the CCM than the previous ones which were made on the one-dimensional Hubbard model on small clusters. We studied the doped 2D Hubbard model at closed shell fillings on finite size square lattices up to the $8 \times 8$ lattice by using the CCM. The total energies were compared with those obtained by using the projector auxiliary field quantum monte carlo (PAFQMC) method and the adaptive sampling quantum monte carlo (ASQMC) method. (An augmented version of the PAFQMC method which reduces the difficulty of the negative sign problem.) We have studied how the accuracy of the CCM depend on the Fermi degeneracy or magnitudes of the gaps of the system to be studied as well as magnitudes of Coulomb interactions and the electron fillings. The former was not studied in the previous work, but it is very crucial if one tries to use the CCM for solid state electrons.

We have studied the 2D Hubbard model: $H = -t \sum_{\langle i,j \rangle, \sigma} (c_i^\dagger c_j^\sigma + H.C.) + U \sum_n n_{i\uparrow} n_{i\downarrow}$. We put $t = 1$.

In the CCM, the exact totally symmetric nondegenerate ground state wavefunction $|\Psi\rangle$ is expressed on the basis of the exponential Ansatz:

$$|\Psi\rangle = \exp(T)|\Phi\rangle,$$

$T = \sum_n T_n$, where $T_n$ is a n-body excitation cluster operator. If $\Phi$ is the Hartree-Fock (HF) wavefunction obtained in the Hartree-Fock limit (easily obtained for a lattice Fermion model), we may neglect the single excitation cluster $T_1$. The double excitation cluster may then be sufficient: $T \approx T_2$. This is an approximation called the coupled-cluster-double (CCD) approximation. Further incorporation of electron correlation is possible by taking into account of the remaining clusters $T_3, T_4, \cdots$. It should be noted however that owing to the exponential form: $\exp(T)$, higher excited configurations than doubly excited configuration, such as quadruple excitation etc., are taken into account in the CCD approximation. If we use the normal product form of the Hamiltonian $H_N$ defined in time-independent perturbation theory, the Schrödinger equation may be written as follows:

$$(H_N - \Delta E) \exp(T)|\Phi\rangle = 0,$$

where $\Delta E = E - E_{HF}$ is the difference of the exact ground state energy and the Hartree-Fock energy. The
Schrödinger equation is solved by multiplying $\exp(-T)$ from the left hand side of the former equation and projecting the resultant equation on excited configurations made from $\Phi$:

$$\langle \Phi^b_e | [H_N \exp(T)]_C | \Phi \rangle = 0,$$

where the subscript C indicates that only connected diagrams are taken into account. The total energy $E$ is given by:

$$E = E_{HF} + \langle \Phi | [H_N \exp(T)]_C | \Phi \rangle.$$

The former equations are the coupled-cluster equations and they and the latter equation constitute a sufficient condition to the Schrödinger equation in the projected space. The advantage of the CCM is that $\exp(T)$ is exactly manipulated without artificial truncations owing to the presence of the subscript C, unlike other variational theories. This guarantees exactly the size consistency condition, which is difficult to satisfy in variational theories. However, there is no variational upper bound given by the CCM energy. The double excitation cluster is expressed as:

$$T_2 = 1/4 \sum_{i,j,a,b} t_{ij}^a t_{ij}^b a_i^\dagger b_j^\dagger,$$

where $i$ and $j$ are annihilation operators for single electron states occupied in $\Phi$ and $a_i^\dagger$ and $b_j^\dagger$ are creation operators for single electron states unoccupied in $\Phi$. The CCD coupled cluster equation is given by:

$$\langle \Phi^b_e | [H_N]_C | \Phi \rangle = \langle \Phi | [H_N]_C | \Phi \rangle + 1/2 \sum_{cd} \langle ab | cd \rangle t_{ij}^{cd} + 1/2 \sum_{kl} \langle kl | \epsilon_k - \epsilon_l \rangle t_{ij}^{kl} - \langle \Phi | [H_N]_C | \Phi \rangle.$$

The total correlation energy is given by:

$$\Delta E = 1/4 \sum_{i,j,a,b} \langle \Phi | (t_{ij}^{a,b} + t_{ij}^{b,a} - t_{ij}^{a,a} - t_{ij}^{b,b}) | \Phi \rangle,$$

$$\langle i||j \rangle$$

and $\epsilon_i$ is the anti-symmetrized two-electron integral defined by: $\langle i||ab \rangle = <i||ab> - \langle i||ba \rangle$. $\epsilon_i$ is the $i$-th single particle energy. Each term in the CCD coupled-cluster equations can be assigned to a diagram. It is known based on diagram theoretical arguments that the effect of the quadruple cluster $T_4$ may be approximately taken into account by neglecting the 8, 9, 13, and 14-th terms in the right hand side of the CCD coupled-cluster equations. Such an approximation is known as the approximate coupled pair (ACP) approximation.

We have calculated the total energy of the 2D Hubbard model by using the ACP approximation. We have compared the ACP energies with exact energies obtained by using the PAFQMC and/or ASQMC methods. These are simply denoted by the QMC method, hereafter. The calculations were made on the $4 \times 4$, $6 \times 6$, and $8 \times 8$ lattices. Electron fillings $\rho = N_e/N_s$ are such that electrons form closed shells on these finite size lattices, where $N_e$ is the number of electrons and $N_s$ is the number of sites. We did not use the second order perturbative selection rules frequently used in $ab$-initio quantum chemistry calculations, which raise systematic errors in results when $U/t$ is large. We have improved an existing fast algorithm by utilizing symmetries of two-electron integrals in the Hubbard model. It takes about 10 minutes of CPU time for a calculation of the $8 \times 8$ lattice on the Alpha workstation with the Alpha 21164/533 MHz CPU chip when we use the ACP approximation. It would not be difficult to do a CCM calculation even beyond a 128 site lattice. We have not done this because the QMC calculations take much more CPU time than the ACP calculations.

The ground state energies of the 2D Hubbard model on the $4 \times 4$ lattice calculated with various methods are plotted in Fig. 1. The electron filling $\rho$ is 0.625. The Restricted Hartree-Fock (RHF) method gives very poor values of the ground state energy even for very small values of $U/t$. The second order Moller-Plesset (MP2) approximation works fairly well in the small $U/t$ region ($U/t \sim 1$), but it fails in the intermediate ($U/t \sim 4$) and the large $U/t$ regions ($U/t \sim 8$). The results obtained by using the ACP approximation are in good agreement with the QMC energies up to the intermediate $U/t$ region on this lattice. The similar results obtained on the $8 \times 8$ lattice are plotted in Fig. 2. $\rho$ is 0.78125. Again, the RHF method is a very poor approximation even for very small value of $U/t$. The MP2 approximation works only in the small $U/t$ region. The ACP approximation works well up to the intermediate $U/t$ region. The deviation of
the energy calculated by using the ACP approximation from the energy calculated by using the QMC method on the $8 \times 8$ lattice is somewhat larger than that obtained on the $4 \times 4$ lattice when $U/t = 8$.

The errors $\%$ of the ACP approximation defined by $(-E_{ACP} + E_{QMC})/E_{QMC} \times 100$ calculated on $4 \times 4$, $6 \times 6$, and $8 \times 8$ lattices are plotted against $U/t$ in Fig. 2. $\rho$ of the $4 \times 4$, $6 \times 6$, and $8 \times 8$ lattices are 0.6250, 0.7220, and 0.78125, respectively. In all cases studied, the errors are less than 1%, when $U/t \leq 4$. The errors grow as we increase $U/t$ and they are larger than 2% when $U/t = 8$. The error in the $8 \times 8$ lattice reaches almost 7% when $U/t = 8$. Thus the ACP approximation breaks down in the large $U/t$ region. The errors increase most rapidly in the case of the $8 \times 8$ lattice. The enhancement of the error in the larger lattice may be mostly due to the difference of $\rho$. $\rho$ is closest to 1 in the $8 \times 8$ lattice in our cases. However, it is interesting to ask whether or not the enhancement is brought about by the decrease of the gap between the lowest unoccupied level (LUL) and the highest occupied level (HOL) in the finite size cluster.

To clarify this point, we study the following generalization Hubbard model: $H = -t \sum_{\langle i,j \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + H.C.) + U \sum_i n_{i\uparrow} n_{i\downarrow} - t' \sum_{\langle i,j \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + H.C.) - t'' \sum_{\langle i,j \rangle} (c_{i\sigma}^\dagger c_{j\sigma} + H.C.)$, where $(i,j)$ and $[i,j]$ denote the second and third nearest pairs of sites on the square lattice, respectively. We put $t = 1$, $t' = -0.2$, and $U = 4$. $t''$ is a variable. We introduced anisotropy $\pm 0.0001$ on the $x$ and $y$ components of $t$ and $t''$, as well as on the $(1,1)$ and $(1,-1)$ components of $t'$. We studied the $8 \times 8$ lattice system with 78 electrons. If $0.0035 \leq t''$ the wave number of the HOL is $\pi, \pi/4$ and the wave number of the LUL is $\pi/2, \pi/2$. The energy separation $\Delta \epsilon \equiv \epsilon(\pi/2, \pi/2) - \epsilon(\pi, \pi/4)$ is a monotonically increasing function of $t''$. We plot the errors $\%$ of the ACP approximation $(-E_{ACP} + E_{QMC})/E_{QMC} \times 100$ as a function of $\Delta \epsilon$ in Fig. 3. The error is almost independent of $\Delta \epsilon$ and is always less than 1%, as long as the CCM equations converge. The CCD equations with the ACP approximation do not converge when $\Delta \epsilon \leq 0.18$. This $\Delta \epsilon$ dependence of the errors indicates the enhancements of the errors in the larger lattices observed in Fig. 2, which comes from the difference of the electron fillings $\rho$. The CCM is less accurate close to the half-filling.

While it seems the ACP approximation works nicely in the Hubbard model up to the intermediate $U/t$ region, it fails in the large $U/t$ region and/or close to half-filling in the two dimension. The $U/t$ dependence of the error was also observed in one-dimensional Hubbard model on small clusters. Direct incorporation of $T_n$, $n \geq 3$ may be necessary in the large $U/t$ region and close to the half-filling. The point in this article is that the accuracy of the CCM does not depend much on the LUL-HOL gap $\Delta \epsilon$. When we apply the crystal orbital method to solid electrons, we use a finite number of $k$ points for $k$ space integrations and constructing excited configurations. Hence, the Fermi degeneracy in metals is replaced by a pseudo-degeneracy among some of the $k$ points. The situation is similar to atomic cluster calculations. In numerical calculations, there are implicitly gaps due to the finite number of the $k$ points, even for metallic electrons.
as well as for electrons in semiconductors and insulators. The fact that the accuracy of the CCM does not depend on such gaps is very favorable, unless numerical results are very dependent on details of the calculation parameters such as increments of $k$ rather than on physical parameters. Hence, the CCM may be successfully applied to ab-initio band calculations on metals as well as semiconductors and insulators.

In conclusion, we have studied the electron correlation problem in the doped 2D Hubbard model with the ACP approximation to investigate whether or not the approximation can be applied to correct the independent particle approximations really used in ab-initio band calculations. We found most of correlation energies of the 2D Hubbard model in the weak and the intermediate $U/t$ region can be accounted for by the ACP approximations. In the large $U/t$ region and/or close to the half-filling, the error of the ACP approximation is not negligible. Further incorporation of the electron correlation effect may be necessary there. The accuracy of the CCM does not depend on the LUL-HOL gap. Hence, the CCM may be successfully applied to ab-initio band calculations on metals as well as semiconductors and insulators.

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