MAGNETIC PHASE TRANSITION IN $CeNi_xCu_{1-x}$ ALLOYS: A KONDO MODEL STUDY

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ABSTRACT

The Kondo Lattice Model (KLM) is one of the most important models for studying heavy-fermion (HF) systems. The KLM has been extensively studied using Cerium base alloys: $CeNi_xCu_{1-x}$. $CeNi_xCu_{1-x}$ alloys are interesting cases due to their magnetic ordering at temperature variations. The introduction of the on-site coulombic interaction term, U, between the conduction electrons is very significant as it comes to play in the various level of concentrations of the Ni and Cu elements in the $CeNi_xCu_{1-x}$ alloys.

The U is a driving force in the transition from the paramagnetic (PM) to the ferromagnetic (FM) state of the KLM. Hence, the Kondo Lattice Model with coulombic interaction (KLMC) between the conduction electrons was considered here using the $CeNi_xCu_{1-x}$ alloys (as a model) and the exact diagonalization technique. An increase in U between the conduction electrons simulates the operation of the increasing Kondo temperature, T_k, as in Coqblin's work.

KEYWORDS: Paramagnetism, Ferromagnetism, Ground State energy, Transition Point.

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1 INTRODUCTION

The Heavy Fermion (HF) materials which are typical examples of strongly correlated systems are materials in which the conduction electrons mix with the almost localized 4f or 5f electrons, and form the strongly renormalized quasiparticles, which have effective masses of 100 – 1000 times larger than the bare value. This strong renormalization is mainly due to the local-kondo-type process, which should be suppressed if an energy gap opens at the Fermi level [1]. One of the canonical models for the description of HF systems is the kondo lattice models [2]. In the regular kondo problem, the spin S_f = ½ of the localized 4f electron (or hole) corresponding to the 4f (or $4f^{13}$) configuration is completely screened at very low temperatures by the spin S_c = ½ of the conduction electrons [3]. The interplay of strong correlation has been observed experimentally in cerium alloys such as Ce(Ni,Cu) or Ce(Pd,Rh). In the Ce(Ni,Cu) alloys with a Cu concentration x, experimental studies have shown magnetic transition with decreasing temperature from being nonmagnetic to paramagnetic and finally to ferromagnetic as temperature is reduced further [4].

An extensive experimental work has been done to study some of the most interesting Cerium alloys. The most studied case is the $CeNi_xCu_{1-x}$ alloys, but we can cite the $CeAu_{1-x}Co_xSi_3$ and $CeRh_xPd_{1-x}$ alloys. The $CeNi_xCu_{1-x}$ alloys were extensively studied firstly, by bulk experimental methods and then by microscopic measurements like neutron diffraction. These alloys are at low temperatures paramagnetic for low Cu concentration (x large). It was firstly shown that, when temperature decreases there are successively a PM phase and then a FM phase [4].

The $CeNi_xCu_{1-x}$ alloy is a heavy fermion (HF) system, and hence can be modeled by the KLM. For large ferromagnetic kondo exchange coupling $J<0$, a FM ground state is obtained with decreasing temperature. This model improves the theoretical description of the kondo systems by providing a simple approach for further calculations of magnetic clusters and can, therefore, account for recent experimental data on Cerium systems [5].

The coulombic interaction between the conduction electrons is the key player in the magnetic phase transitions of alloys such as Ce(Ni,Cu). The physical operations of the Kondo temperature T_k, tends to affect the activities of the conduction electrons.

The transition from a PM phase to a FM phase observed with decreasing temperature in $CeNi_xCu_{1-x}$ alloy is a complicated problem which needs a description of its properties and their evolution with temperature. By increasing the conduction electrons, the low temperature transition can be improved upon to have transition

at higher temperature. In this paper, a systematic study of the KLMC is undertaken by the exact diagonalization technique.

2 METHODOLOGY

The conventional KLM Hamiltonian, H, given by Tsunetsugu [3] is

$$H = -t\sum_{i\sigma}\left(C_{i\sigma}^{+}C_{i+1\sigma} + H.C.\right) + J\sum_{i} S_{if}.S_{ic} \qquad \text{.....(1)}$$

where in eqn. (1) the hopping integral parameter, t (the t term of the Hamiltonian) describes the itinerancy tendency of electrons, H.C. is the Hermitian Conjugation of $C_{i\sigma}^{+}C_{i+1\sigma}$,

and

$$S_{if} = \sum_{\alpha\beta}\frac{1}{2} f_{i\alpha}^{+}\sigma_{\alpha\beta}^{\mu} f_{i\beta}$$

$$S_{ic} = \sum_{\alpha\beta}\frac{1}{2} C_{i\alpha}^{+}\sigma_{\alpha\beta}^{\mu} C_{i\beta}$$

Despite the enormous effort put in by several researchers to describe the ground state of the KLM, only little have been achieved [5]. For example, the ferromagnetic ground state of the KLM has not been fully understood. This phenomenon has to do with the coulombic interaction that exists between the conduction electrons. Extensions of the actual KLM given by equation (1), have to be done with the inclusion of the on-site coulombic interaction term, U. With the inclusion of the U term, the Hamiltonian (1) becomes

$$H = -t\sum_{i\sigma}\left(C_{i\sigma}^{+}C_{i+1\sigma} + H.C\right) + J\sum_{i} S_{if}.S_{ic} + U\sum_{i}\left(C_{i\uparrow}^{+}C_{i\uparrow} - \frac{1}{2}\right)\left(C_{i\downarrow}^{+}C_{i\downarrow} - \frac{1}{2}\right) \qquad \text{.....(2)}$$

Equation (2) is known as the kondo lattice model with coulombic interaction between the conduction electrons (KLMC) [7].

The KLMC Hamiltonian was used to investigate the ground state properties in the lattices studied in this paper. With these tools, it is possible to study the KLMC systems of 2 electrons on 2 sites and 2 electrons on 3 sites.

3 CALCULATIONS

Results for a system of 2 electrons on a 2- site (1-D) lattice system using the kondo lattice model with coulombic interaction between conduction electrons (KLMC) are given below.

For two electrons on two (2) sites we have six states i.e

$S = {}^{2N}C_n = {}^{4}C_2$

$$S = \frac{(2N)!}{n!(2N-n)!} \quad where$$

N = number of site = 2

n= number of electrons = 2

$$S = \frac{(2x2)!}{2!(2x2-2)!} = 6 \quad \text{basis states}$$

Generating the states, we have explicitly

$$\left.\begin{aligned} |1\rangle &= |1\uparrow 1\downarrow\rangle \\ |2\rangle &= |2\uparrow 2\downarrow\rangle \\ |3\rangle &= |1\uparrow 2\downarrow\rangle \\ |4\rangle &= |1\downarrow 2\uparrow\rangle \\ |5\rangle &= |1\uparrow 2\uparrow\rangle \\ |6\rangle &= |1\downarrow 2\downarrow\rangle \end{aligned}\right\} \quad . \qquad \ldots\ldots(3)$$

Using the Hamiltonian (1) to act on the states (3) the following were obtained;

$$\left.\begin{aligned}
H|1\rangle &= t|4\rangle - t|3\rangle \\
H|2\rangle &= t|4\rangle - t|3\rangle \\
H|3\rangle &= -t|1\rangle - t|2\rangle + \frac{3}{2}J|3\rangle - U|3\rangle \\
H|4\rangle &= t|1\rangle + t|2\rangle + \frac{3}{2}J|4\rangle - U\ |4\rangle \\
H|5\rangle &= \frac{3}{2}J|5\rangle - U|5\rangle \\
H|6\rangle &= \frac{3}{2}J|6\rangle - U|6\rangle
\end{aligned}\right\} \quad \text{.....(4)}$$

The ground state energies for both the singlet E_s and the triplet E_t are given by (5) and (6) respectively.

$$E_s = \frac{1}{4}\left(3J - \sqrt{64t^2 + (3J - 2U)^2} - 2\mathrm{U}\right) \quad \text{...(5)}$$

$$E_t = \frac{3J}{2} - 2U \quad \text{. . .(6)}$$

[†] The results for a system of 2 electrons on 3- sites (1-D), 3 electrons on 3- sites (1-D), 2 electrons on a 4- site (1-D) and 4 electrons on 4- sites (1-D) lattice system using the kondo lattice model with coulombic interaction between conduction electrons (KLMC) are given below.

The ground state energies for both the singlet E_s and the triplet E_t for 2 electrons on 3-sites (1-D) are given by (7) and (8) respectively. This is obtained numerically for t=J=U=1.

$$E_s = 0.000 \quad \text{...(7)}$$

$$E_t = 0.086 \quad \text{...(8)}$$

The ground state energies for both the singlet E_s and the triplet E_t for 3 electrons on 3- sites (1-D) are given by (9) and (10) respectively. This is obtained numerically for t=J=U=1.

$$E_s = -0.164 \quad \text{...(9)}$$

$$E_t = 1.750 \quad \text{..(10)}$$

The ground state energies for both the singlet E_s and the triplet E_t for 2 electrons on a 4- site (1-D) are given by (11) and (12) respectively. This is obtained numerically for t=J=U=1.

$$E_s = 0.000 \quad \text{.(11)}$$

$$E_t = 1.736 \quad \text{.(12)}$$

The ground state energies for both the singlet E_s and the triplet E_t for 4 electrons on 4- sites (1-D) are given by (13) and (14) respectively. This is obtained numerically for t=J=U=1.

$$E_s = 0.001153 \quad \text{...(13)}$$

$$E_t = -0.148123 \quad .(14)$$

4 DISCUSSION OF RESULTS

It was observed from Table 1 and Figure 1, that as the value of the coulombic interaction, U, is varied between U=1.5 and U = 6.0, keeping t and J, constant, the FM phase became stable.

From Table 2 and Figure 2, it was observed that keeping t = J=1 constant, as $U \rightarrow +\infty$, the lattice transits from a PM phase to a FM phase. This was achieved when U is gradually increased from U = 1.00 to U = 10.00.

It was also observed from Table 3 and Figure 3 that as the coulombic interaction term, U, increases, the PM phase gradually becomes unstable. At point U = 1.9, there was a transition from a PM phase to a FM phase.

It was also observed in Table 4 and Figure 4 that the presence of U suppresses the PM phase and greatly favors the FM phase. As the value of U increases from U = 1.0 to U = 1.9, a transition from PM phase to FM phase was observed.

Finally, it was observed in Table 5 and Figure 5 that as U, the coulombic interaction term, is gradually increased from U = 1.00 to U = 5.00, the FM ground state becomes more stable. This was achieved as J = t =1 are kept constant.

Thus, the FM phase stabilizes as U is increased.The reverse would be the case if U decreases and t increases. An electronic charge in motion gives rise to a tiny magnetic field. Each electronic charge in motion always feel the presence of other neighbouring electrons because of the Coulombic and spin interactions between them. The resulting interaction of these charges in motion is what is usually referred to as *electron correlations.*

At half-filling, i.e. for 2 electrons on 2-sites, 3 electrons on 3-sites and 4 electrons on 4-sites the KLMC exists in an unstable PM ground state for J<0 and it undergoes a transition to a FM ground state (triplet ground state) as U increases, i.e., as $U \rightarrow +\infty$.

For the case of slightly away from half filling (2 electrons on 3 sites) and quarter filling (2 electrons on 4-sites) for the KLMC, the effect of increasing, U, (i.e., as $U \rightarrow +\infty$) helps to stabilize the FM ground state.

The $CeNi_xCu_{1-x}$ alloy is a heavy fermion (HF) system, and hence can be modeled by the KLMC. These alloys are at low temperatures paramagnetic for low Cu concentration (x large). The transition observed with decreasing temperature from a PM phase to a FM phase is a complicated problem which needs a description of its properties and their evolution with temperature. The role of the Kondo temperature,T_k in the $CeNi_xCu_{1-x}$ alloy was investigation also using the neutron diffraction technique [5]. This has shown that FM transition is feasible for a stable PM state of the $CeNi_xCu_{1-x}$ alloy with increasing T_k. This is analogous to the theoretical parameter U in the KLMC. At any particular concentration parameter (x), a transition temperature (T_k) is found. A decrease in x (i.e. increase in conduction electrons) results in an increase in the transition temperature, due to the increase in coulombic interaction U between the conduction electrons. So, more conduction electrons, more coulombic interactions and an increase in T_k . As the temperature decreases, atomic vibrations become small and on-site coulombic interaction U between the conduction electrons increases as the electrons can travel through a metallic crystal more easily when the vibrations of the atoms are small, hence, decreasing the temperature leads to magnetic ordering. There is a dependence of T_k on concentration parameter x. An increase in U is analogous to an increase in T_k. The coulombic interaction U is dependent on the magnitude of J. For $J \rightarrow -\infty$ the PM state is unstable for small perturbation of U [7].

5 CONCLUSIONS

In conclusion, the exact diagonalisation method has been employed to provide information on the behavior of two interacting electrons on 1-D lattices. Finite sized lattices with periodic boundary conditions were specifically considered and the dynamics of the interacting electrons was described by the KLMC.

The results obtained in the KLMC are in agreement with the experimental work of [1] who experimentally worked on the Cerium alloy, $CeNi_xCu_{1-x}$, using neutron diffraction. He studied the experimental situation of x between 0.3 and 0.7. The experimental magnetic phase reveals a transition from the PM state to the FM phase. An increase in U in the KLMC is analogous to an increase in T_k.

The coulombic interaction U in KLMC has been used to throw more light on the anomalous behaviour exhibited by $CeNi_xCu_{1-x}$ alloy. The anomalous behaviour is such that at high temperature (above 100k) the $CeNi_xCu_{1-x}$ alloy which is supposed to be magnetic exists in a nonmagnetic state. There is a sudden appearance of magnetic properties (PM phase) as temperature is gradually reduced. This is explained considering the fact that at low temperature there is reduced atomic vibration and the electrons can move more easily thereby increasing the coulombic interaction, U between them.

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TABLE 1:The lowest energies of both the singlet and the triplet states obtained numerically as the columbic interaction term , U, is varied while the exchange interaction term, J, and t are kept constant , (i.e., J=-5, and t=1). The E_t forms the ground state energy

t (the hopping integal)	J (the exchange interaction term)	U (the coulombic interaction term)	E_s (the singlet state energy)	E_t (the triplet state energy)
1.00	-5.00	1.50	-9.42	-10.50
1.00	-5.00	2.00	-9.90	-11.50
1.00	-5.00	2.50	-10.38	-12.50
1.00	-5.00	3.00	-10.86	-13.50
1.00	-5.00	3.50	-11.35	-14.50
1.00	-5.00	4.00	-11.83	-15.50
1.00	-5.00	4.50	-12.32	-16.50
1.00	-5.00	5.00	-12.81	-17.50
1.00	-5.00	5.50	-13.30	-18.50
1.00	-5.00	6.00	-13.79	-19.50

TABLE 2: The lowest energies of both the Singlet and the triplet states obtained numerically as the coulombic interaction term, U, is varied with other parameters remaining constant (i.e., J=1 and t=1). The E_s forms the initial ground state energy.

t (the hopping integral)	J (the exchange interaction term)	U (the coulombic interaction term)	E_s (the singlet state energy)	E_t (the triplet state energy)
1.00	1.00	1.00	0.00	0.09
1.00	1.00	2.00	0.00	0.91
1.00	1.00	3.00	0.00	-0.07
1.00	1.00	4.00	0.00	-1.08
1.00	1.00	5.00	0.00	-2.08
1.00	1.00	6.00	0.00	-3.08
1.00	1.00	7.00	0.00	-4.08
1.00	1.00	8.00	0.00	-5.08
1.00	1.00	9.00	0.00	-6.08
1.00	1.00	10.00	0.00	-7.08

Table 3 : The lowest energies of both the singlet and the triplet states obtained numerically as the coulombic interaction term, U, is varied and other parameters remaining constant i.e.,J=1 and t=3

t (the hopping integral)	J (the exchange interaction term)	U (the coulombic interaction term)	E_s (the singlet state energy)	E_t (the triplet state energy)
3.00	1.00	1.200	-1.147	0.450
3.00	1.00	1.300	-1.093	0.300
3.00	1.00	1.400	-1.038	0.150
3.00	1.00	1.500	-0.982	0.000
3.00	1.00	1.600	-0.923	-0.150
3.00	1.00	1.700	-0.862	-0.300
3.00	1.00	1.800	-0.799	-0.450
3.00	1.00	1.900	-0.733	-0.731
3.00	1.00	2.000	-0.664	-0.750
3.00	1.00	2.100	-0.591	-0.900

Table 4 : The lowest energies of both the singlet and the triplet states obtained numerically as the coulombic interaction term , U, is varied and other parameters remaining constant i.e., t=1 and J=1.

t (the hopping integral)	J (the exchange interaction term)	U (the coulombic interaction term)	E_s (the singlet state energy)	E_t (the triplet state energy)
1.00	**1.00**	**1.00**	**0.00**	**1.73**
1.00	**1.00**	**1.10**	**0.00**	**1.53**
1.00	**1.00**	**1.20**	**0.00**	**1.33**
1.00	**1.00**	**1.30**	**0.00**	**1.13**
1.00	**1.00**	**1.40**	**0.00**	**0.93**
1.00	**1.00**	**1.50**	**0.00**	**0.73**
1.00	**1.00**	**1.60**	**0.00**	**0.53**
1.00	**1.00**	**1.70**	**0.00**	**0.33**
1.00	**1.00**	**1.80**	**0.00**	**0.13**
1.00	**1.00**	**1.90**	**0.00**	**-0.02**

Table 5 : The lowest energies of both the singlet and the triplet states obtained numerically as the coulombic interaction term , U, is varied and other parameters remaining constant i.e., t=J=1.

t (the hopping integral)	J (the exchange interaction term)	U (the coulombic interaction term)	E_s (the singlet state energy)	E_t (the triplet state energy)
1.00	**1.00**	**1.00**	**0.001**	**-0.148**
1.00	**1.00**	**2.00**	**0.169**	**-0.023**
1.00	**1.00**	**3.00**	**0.065**	**-0.030**
1.00	**1.00**	**4.00**	**-0.133**	**-0.392**
1.00	**1.00**	**5.00**	**-0.137**	**-0.402**

Fig 1: Lowest energies plotted against U for a system of 2 electrons on 2 Sites (KLMC).

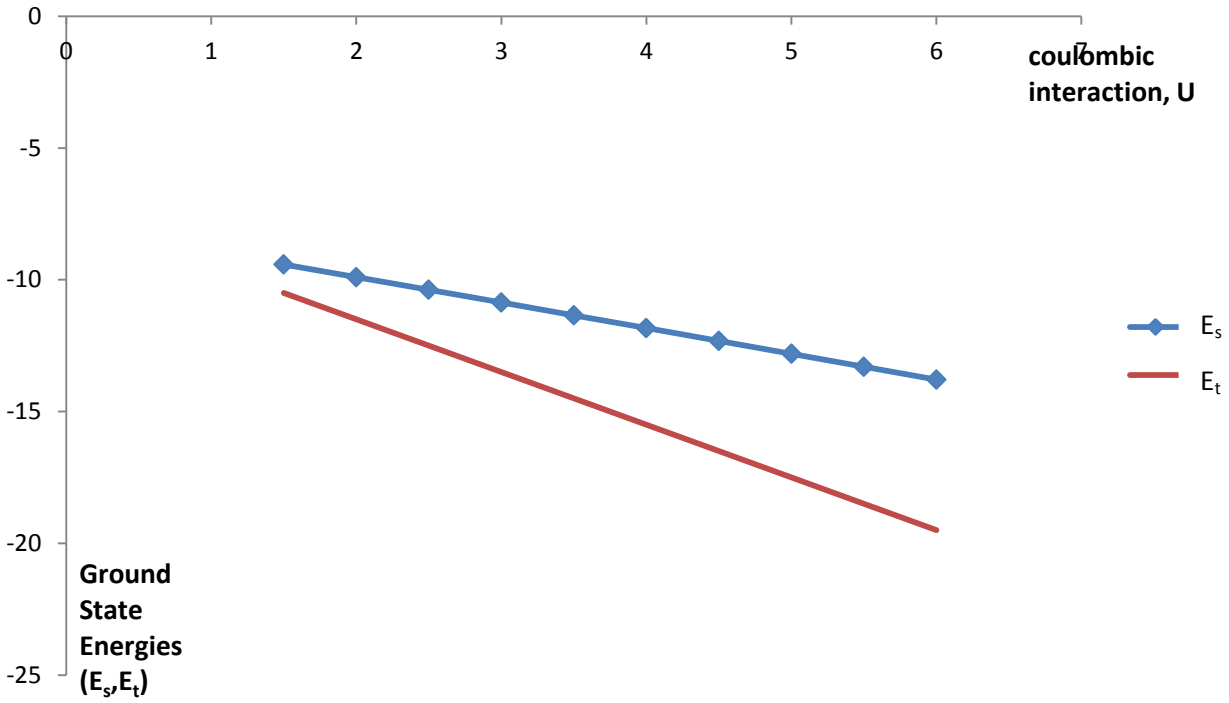

Fig 2: Lowest energies plotted against U for a system of 2 electrons on 3 Sites (KLMC).

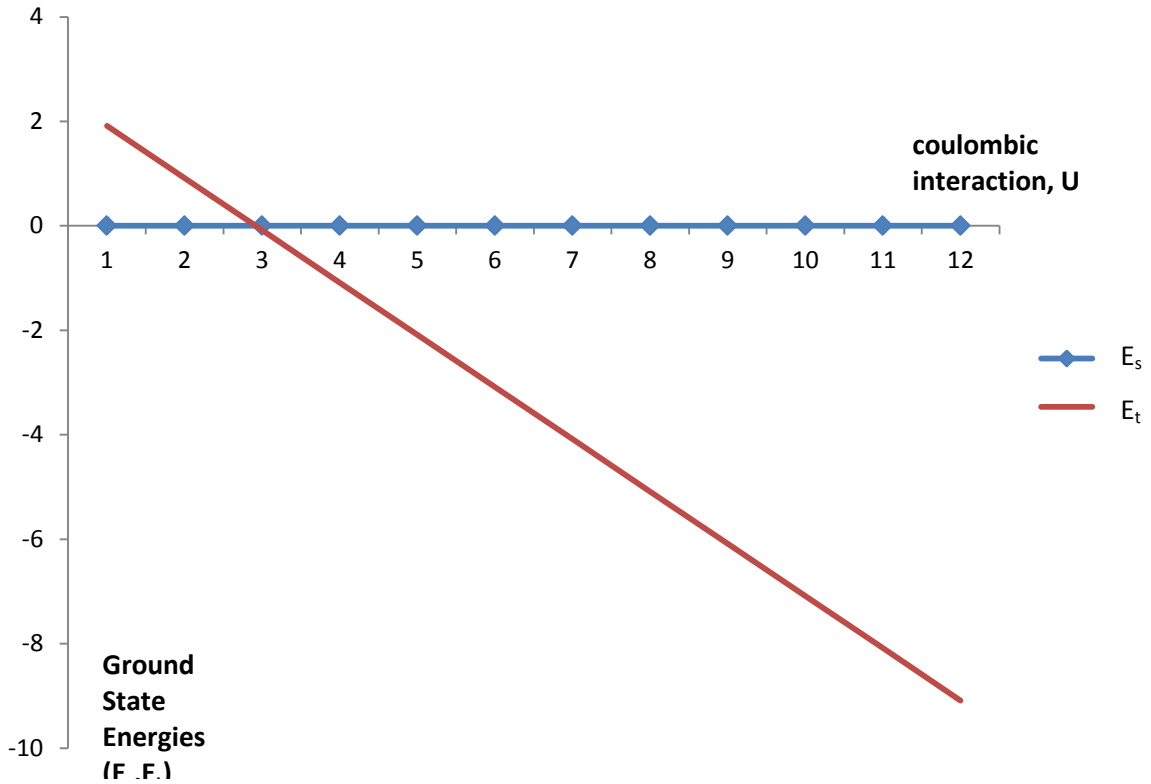

Fig 3: Lowest energies plotted against U for a system of 3 electrons on 3 Sites (KLMC).

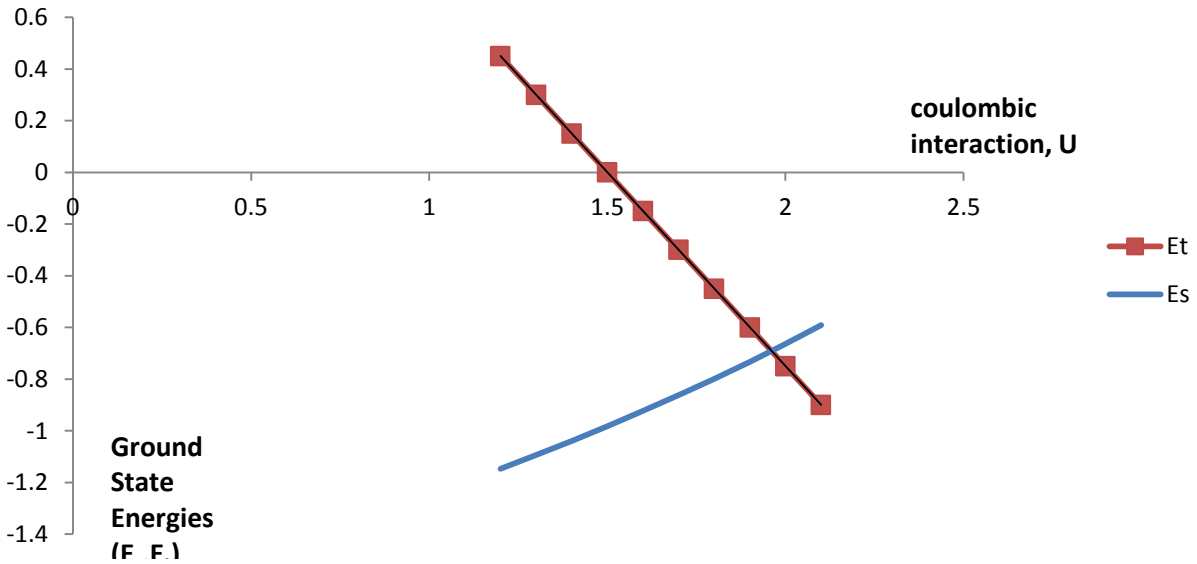

Fig 4: Lowest energies plotted against U for a system of 2 electrons on 4 Sites (KLMC).

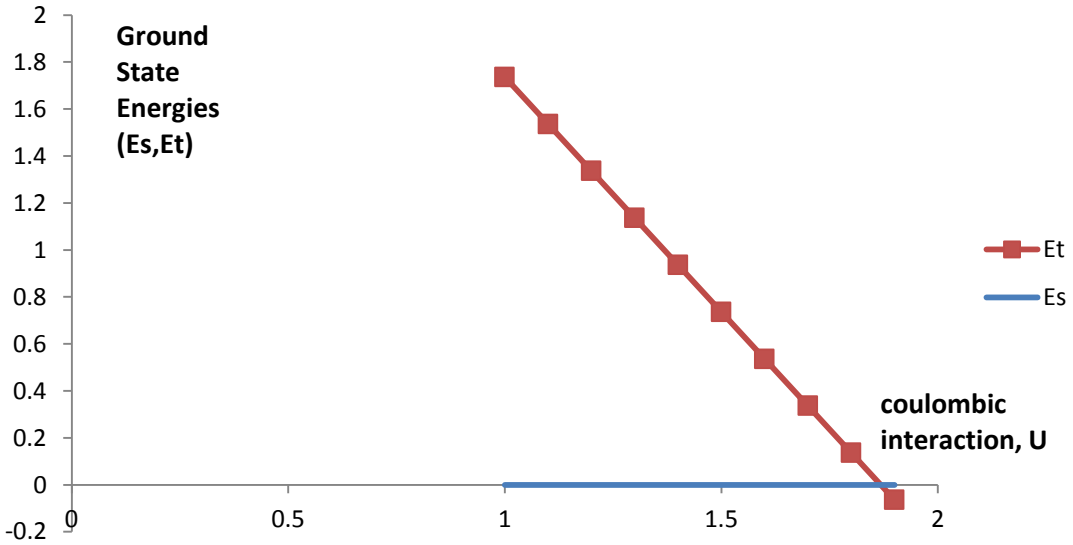

Fig 1: Lowest energies plotted against U for a system of 4 electrons on 4 Sites (KLMC).