Origin of Non-Fermi Liquid Behavior in Heavy Fermion Systems: A Conceptual View

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Abstract

We critically examine the non-Fermi liquid (NFL) behavior observed in heavy fermion systems located close to a magnetic instability and suggest a conceptual advance in physics in order to explain its origin. We argue that the treatment of electronic states responsible for magnetism near the Quantum Critical Point (QCP), cannot be accomplished using the present-day theory treating electronic exchanges within the quantum mechanical formalism; instead new ideas need to be introduced in it while treating these electronic states. The observed NFL behavior can be explained within such a scenario. As a sequel we attempt to discuss its consequences for the explanation of high-T_C superconductivity observed in Cuprates.

Keywords: Unconventional Superconductivity, Fermionic Exchange Mechanism, Kondo Effect

I. INTRODUCTION

NFL behavior is observed in heavy fermion systems when the system is tuned towards a magnetic instability known as the QCP [1, 2]. Far away from QCP (but close to 0 K) one observes normal Fermi liquid behavior as expected for metals by Landau [3]. Landau's Fermi liquid theory is remarkably successful in explaining the low temperature behavior of paramagnetic heavy fermion systems despite the strength of electronic correlations being as large as 6-7 eV when described within the Anderson model [4]. A big question is: What causes the breakdown of Landau's Fermi liquid theory close to the QCP while it remains valid for various ground states far away from it. This has been a long standing problem within the scientific community and extensive experimental and theoretical investigations have been performed in order to shed light on it. Many theoretical progresses have been made in order to explain NFL behavior e.g. models based on multichannel Kondo effect, models based on QCP, models based on disorder etc. [1, 2]. Despite the intense study, a consensus over the origin of NFL behavior has not been reached yet.

In this paper, we propose a conceptual advance in physics over the treatment of electronic states close to the QCP. Our picture is based on certain overlooked issues in quantum mechanics which led to improper theoretical treatment of 'magnetic' states close to the QCP. These issues, when considered, readily explain the observed NFL behavior.

II. RESULTS AND DISCUSSION

In order to illustrate our point, we take recourse to the single ion Kondo problem [5]. Let us assume a two electron system: a 'magnetic' 4*f* electron and an itinerant valence electron. The ground state for the single ion Kondo problem is the non-magnetic Kondo singlet state represented by $|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v$. This quantum mechanical state describes indistinguishable particles as is evident from its exchange symmetry. In this state the 4*f* electronic degrees of freedom have to be included in the Fermi volume and hence the 4*f* electrons are fermions just like the valence electrons (fermionic and bosonic wave functions necessarily need to obey certain exchange symmetries as demanded by quantum field theory). Since this is a singlet state it has no net magnetic moment. The 4*f* electron's magnetic moment has been compensated by its anti-ferromagnetic coupling to the spins of the valence electrons. A question is: How to describe the two electron state at higher temperatures. At high temperatures the 4*f* electrons recover their localized magnetic moments and hence a singlet description is definitely not appropriate. Within the present-day quantum mechanical formalism, the low to high temperature transition in single ion Kondo systems can be modeled by a singlet to triplet transition wherein the high temperature state is the triplet state which has a magnetic moment [6, 7]. Such a treatment might explain the recovery of 4*f* magnetic moment at high temperatures. However within such a treatment the 4*f* electrons continue to remain as 'fermions' (indistinguishable particles) even at higher temperatures on account of their quantum mechanical spin triplet description (obeying fermionic exchange symmetry) thereby participating in the Fermi volume. Therefore, within this picture, the Fermi volume cannot change with temperature.

In case of a Kondo lattice, Kondo phenomenon can be modeled as the coherent screening of the periodic 4*f* electrons by valence electrons creating a coherent Kondo state below the coherence temperature, T_{coh} [8, 9]. The development of Kondo coherence leads to the loss of magnetic moments of the localized 4*f* electrons at low temperatures. The temperature dependent large-small Fermi surface transition in Kondo lattices is a long standing problem in the community. It is believed that the Fermi surface in Kondo systems should expand below T_{coh} in order to incorporate 4*f* electronic degrees of freedom into it as a result of

the Kondo coherence [10-12]. At high temperatures the Fermi surface remains small since the Kondo coherent state is not yet established and the 4*f* electrons are localized; hence they are excluded from the Fermi volume. Experimental evidence for such an expansion has been obtained in the de Haas van Alphen experiments performed on many Kondo lattices at very low temperatures [13]. These results have been successfully interpreted by considering the 4*f* electronic states as being a part of the Fermi volume and hence demonstrate the expanded character of the Fermi surface. The question is how the large-small Fermi surface transition should be modeled theoretically? There are few approaches based on dynamical mean field theory (DMFT) which have attempted to model the large-small Fermi surface transition in $CeIrIn_5$ [14, 15]. In this paper, we present an elegant conceptual picture to model this Fermi surface transition. We provide conditions under which electrons can/cannot be treated as fermions.

Within the present-day quantum mechanical description of the Kondo phenomenon, the Fermi surface topology would undergo restructuring merely without any explicit expansion since the 4*f* electrons remain as 'fermions' at all temperatures (since they are always described in a state obeying the fermionic exchange symmetry at all temperatures) and hence their participation in the Fermi volume cannot vary with temperature; a fact in contrast to experiments which have indeed demonstrated the large-small Fermi surface transition in Kondo lattices explicitly. Hence such a description of the Kondo phenomenon is inappropriate. Instead we propose a new scenario in order to explain the temperature dependent Kondo phenomenon. We claim that the high temperature state in any Kondo model should not be described as a quantum mechanical state demonstrating fermionic exchange symmetry. For example, within the single ion Kondo model the high temperature state corresponding to a localized 4*f* electron should not be described as a spin triplet state but rather should be described as a state like $|\uparrow\rangle_f|\downarrow\rangle_v$ (or $|\downarrow\rangle_f|\uparrow\rangle_v$ equivalently since both states are degenerate in the absence of a magnetic field). The reason why fermionic exchange symmetry should be forbidden for the high temperature state can be understood once we understand the effect of exchange between two electrons on their individual characteristics (see supplementary information section A). Thus this high temperature state cannot have exchange symmetry as required for fermions and therefore does not represent a quantum mechanical state describing indistinguishable particles. Instead, it is a state describing distinguishable particles wherein a particle exchange gives rise to a new state. Therefore we claim that the 4*f* electrons cannot be treated as 'fermions' at high temperatures. Therefore the transition from the state $|\uparrow\rangle_f|\downarrow\rangle_v$ (or $|\downarrow\rangle_f|\uparrow\rangle_v$) at high temperatures to a state $|\uparrow\rangle_f|\downarrow\rangle_v-|\downarrow\rangle_f|\uparrow\rangle_v$ at low temperatures cannot be modeled within the present-day (quantum mechanical) theory of exchange mechanism among electrons and requires introduction of new principles into it (see supplementary information A and C for details). Within this new picture the large-small Fermi surface transition can be explained naturally. The state $|\uparrow\rangle_f|\downarrow\rangle_v-|\downarrow\rangle_f|\uparrow\rangle_v$ at low temperatures describes both 4*f* and valence electrons as fermions participating in the Fermi volume that corresponds to a large Fermi surface while the state $|\uparrow\rangle_f|\downarrow\rangle_v$ (or $|\downarrow\rangle_f|\uparrow\rangle_v$) at high temperatures represents a localized 4*f* electron which is excluded from the Fermi surface the Fermi volume contains only the itinerant valence electron and hence the Fermi surface is small.

This conceptual picture may be easily visualized by comparing spatial extensions of wave functions for the localized 4*f* and itinerant valence electrons. We argue that in order for an electronic system to be considered as a 'fermionic' system and treated within the Landau's Fermi liquid formalism, the spatial distribution of the amplitudes of the wave functions of all the constituent electrons must be identical to each other. Alternately one can state that for a fermionic system the spatial profile of the wave functions of all constituent electrons must be identical to each other. On the contrary when a 4*f* electron is localized then it does not lead to such identical profile since the amplitude of 4*f* electronic wave function peaks in the region of localization and becomes negligible far away from it whereas the amplitude of valence electronic wave function continues to be spatially uniform. Thus there is inconsistency between the spatial profiles of the wave functions of 4*f* and valence electrons giving rise to 'distinguishability' between them. Hence the localized 4*f* electron cannot be treated as a fermion. This happens in Kondo systems at high temperatures when the 4*f* electron localizes. At low temperatures the spatial profile of the 4*f* electronic wave function is identical to that of the valence electronic wave function due to quantum entanglement. Hence they both become indistinguishable and the 4*f* electron is therefore a fermion. A remarkable consequence of this idea can be seen when tuning the ground state of Kondo lattices across QCP. When the Kondo ground state is formed in case of paramagnetic Kondo lattices the Fermi surface is large due to the occupancy of 4*f* electrons in the Fermi volume. The valence band contains both 4*f* and itinerant valence electrons and the Fermi liquid theory holds for low energy excitations of the valence band since both 4*f* and itinerant valence electrons are fermions in this case. On the contrary when the 4*f* electron is completely localized in the magnetically ordered ground state of Kondo lattices then it cannot be treated as a fermion as just discussed above. In this case the Fermi volume contains only the itinerant valence electrons excluding the 4*f* electrons (small Fermi surface). However, surprisingly, the Fermi liquid theory holds true in this case too for the low energy valence band excitations despite the 4*f* electron not being a fermion. The explanation to this mystery is that the 4*f* electrons being completely localized do not participate in the valence band and hence cannot contribute to the low energy excitations of the valence band. The low energy excitations of the valence band result only from itinerant valence electrons which are fermions anyway and hence Fermi liquid theory holds even within the magnetically ordered phase at low temperatures (It is to be noted that the applicability of Fermi liquid theory is always in the context of valence band in solids since it is the valence band which influences the low energy excitations in solids. The 'localized' electrons do not contribute to the valence band and therefore cannot influence the low energy excitations in solids). When the system is tuned towards QCP, the degree of 4*f* electron delocalization increases gradually until we approach close to QCP where we get *comparable* contribution from the 4*f* electron and the itinerant valence electrons in the valence band. Hence the low energy excitations of the

valence band for systems close to QCP are influenced jointly by the 4*f* and itinerant valence electrons. Its effect on the observation of Fermi liquid behavior from the valence band will be explained at the end of next paragraph.

There are two possible theoretical scenarios currently believed for explaining the nature of 4*f* electrons in the vicinity of QCP. In one case it is predicted that the 4*f* magnetic moments are completely screened due to the Kondo effect at QCP and the magnetic order results within the itinerant electron gas [16-18] whereas the other scenario predicts that the 4*f* electrons remain localized even at QCP and the magnetic order results from the localized electrons [19, 20]. All of these predictions are made by theoretical models within the quantum mechanical formalism. Either of these two scenarios have a certain amount of experimental support hence the debate over which among these two scenarios prevail is still ongoing. We provide a radically new theoretical picture in this regard. We argue that since the 4*f* electron transforms from a fermion in the paramagnetic ground state to a non-fermion (a distinguishable particle) in the magnetically ordered state through the QCP, the corresponding theoretical description for the transformation cannot be obtained within the existing quantum mechanical framework handling the exchange mechanism among electrons but requires new principles to be introduced into it. In this context we argue that the 4*f* electrons maintain their partial localized character even at the QCP which forbids their description as 'fermions'. Consequently the valence electronic system close to QCP, containing both the partially localized 4*f* (non-fermion) and itinerant valence electrons (fermion), cannot be modeled within the Landau's Fermi liquid formalism and hence NFL behavior emerges (For another explanation see Supplementary Information Section B).

Furthermore it is observed that the ground state close to QCP in many heavy fermion systems is superconducting in which the superconducting pairing mechanism is different from conventional phonon mediated. It is commonly believed that the unconventional superconducting pairing mechanism is electronic in origin. The question is why such an unconventional pairing mechanism is operative only close to QCP and not far away from it? We attempt to explain this in the following way. It is well known that every electronic system has a tendency to form a bosonic ground state in order to reduce its energy since bosons can condense into the same state at low temperatures whereas the fermions cannot do so due to Pauli's exclusion principle. It is due to this tendency that superconducting ground states are formed in case of certain metallic systems. However one needs a suitable mechanism to form superconducting Cooper pairs (bosons) out of electrons (fermions) in order to realize the superconducting ground state. We claim that the unconventional pairing mechanism is a consequence of the fact that the valence electrons for systems close to QCP are not fermionic in nature. Therefore the tendency to form a bosonic ground state in the valence electronic system is supported in such a case and superconductivity emerges. Far away from QCP (but close to 0K) the valence electrons are fermions and hence the unconventional pairing mechanism is not supported in such a case. Since Fermi liquid theory does not hold close to QCP, quasi-particles do not form and therefore it is possible that the itinerant valence electrons find a glue to bind themselves together into a Cooper pair. Analogous to the case of phonon mediated superconductors where the glue is provided by the electron phonon coupling which causes an effective attractive interaction between two electrons giving rise to Cooper pairs [21, 22], the glue in case of heavy fermion superconductors seems to be generated by the temperature dependent attractive interaction between 4*f* and itinerant valence electron – *Note that Kondo effect is an example of asymptotic freedom [23, 24] in condensed matter in which the strength of attractive coupling between 4f and valence electron is temperature dependent. The strength of this coupling increases with reducing temperature (see Supplementary Information Section C). This coupling is a result of hybridization between 4f and valence electron. An effect of such temperature dependent coupling is clearly visible in the photoemission spectra as demonstrated in our earlier publication [25]* – effectively giving rise to an attractive interaction between two itinerant valence electrons binding them into a Cooper pair.

We anticipate that a similar reasoning holds while explaining the unconventional superconductivity observed in high T_C Cuprate superconductors. Here too the metallic system above the superconducting transition temperature manifests anomalous behavior and therefore is called as a 'strange metal' [26, 27]. It is also believed that a QCP exists deep within the superconducting dome. Therefore it is quite likely that the unconventional pairing mechanism in Cuprate superconductor results from the fact that its valence electronic system is not fermionic in nature similar to the case of heavy fermion superconductors and hence unconventional superconductivity is supported in such a case. It is likely that the valence holes (electrons) in the hole (electron) doped Cuprate superconductors have an attractive temperature dependent coupling (resulting from the hybridization between the valence state and the localized magnetic moment) with the localized magnetic moment of the Cu ion (in d^9 electronic configuration) which gives rise to an effective attractive interaction between two valence holes (electrons) producing Cooper pairs causing high T_C superconductivity — a similar picture as we proposed for explaining heavy fermion superconductivity.

III. CONCLUSIONS

In summary, we argue that the high temperature state in the Kondo model cannot be treated within the existing theory of electronic exchange within the quantum mechanical formalism. Instead we claim that the correct modeling of the Kondo phenomenon would require the theory to be revised and updated along the lines as suggested in this manuscript. This new picture easily explains the experimentally observed large-small Fermi surface transition. The strength of our proposal comes from the fact that we highlight certain overlooked issues in quantum mechanics which led to inappropriate theoretical treatment of electronic states subjected to different degrees of localization. The key conceptual advance provided by this manuscript is the claim that not every electron qualifies to be called as a fermion and can be modeled within Landau's Fermi liquid formalism.

Partially localized electrons alongside itinerant electrons together cannot be treated as a fermionic system and hence it displays NFL behavior in its low energy excitation spectrum. The non-fermionic nature of valence electronic system close to QCP supports the tendency of the electronic system to form a bosonic superconducting ground state and hence unconventional superconductivity is observed at QCP in heavy fermion systems. We project that a similar picture possibly holds for the explanation of unconventional superconductivity observed in high T_C Cuprate superconductors as well. We argue that the Cooper pairing glue in case of heavy fermion or Cuprate superconductors results from the existence of an attractive temperature dependent coupling between valence states and the localized magnetic moment (resulting from the hybridization between the valence state and the localized magnetic moment) which can give rise to an effective attractive interaction between two valence electrons (or holes) forming Cooper pairs yielding unconventional superconductivity. We strongly believe that our manuscript would definitely play an important foundational role for the future development of theoretical models addressing these issues.

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SUPPLEMENTARY INFORMATION

A. Effect of an Electronic Exchange on Their Individual Characteristics

{Note that at the quantum level the word 'identical' particle has been coined for a class of particles which have similar physical characteristics. E.g. any two electrons have same bare mass, same bare charge, same intrinsic spin momentum $(\hbar/2)$ etc. Hence any two electrons in the universe 'apparently' look identical with each other. As a result an exchange between these identical particles is not expected to give rise to a new electronic state/configuration as against the case with the classical particles. Hence these 'identical' particles have been treated as indistinguishable particles as far as their statistical properties are concerned in contrast to distinguishable classical particles. It is due to this fact that their wave functions have to obey fermionic exchange symmetry which ensures that all physical attributes corresponding to their electronic state are invariant upon a particle exchange. This is our current understanding in basic physics. These indistinguishable particles are called as fermions for particles with half-integral spin and bosons for particles with integral spin. So according to our current understanding if you take one electron from the conduction band and another one from a core level inside the solid then the two electron wave function (for both these 'identical' particles) will continue to obey the exchange symmetry as applicable for fermions. Exactly here we claim that such of our prevalent understanding of basic physics is inaccurate. The core electron is localized at a crystal lattice site while the conduction electron is itinerant within the whole of the solid. An exchange between them will give rise to consequences which contradict the localized nature of the core level electron as explained in the following}.

Consider two electrons: electron#1 and electron#2. Suppose they are in states A and B respectively. The fermionic wave function (for indistinguishability between both these electrons) for their two electron state is $|1\rangle_A|2\rangle_B - |2\rangle_A|1\rangle_B$. In this state one can see that electron#1 is in both states A and B simultaneously due to the exchange symmetry as applicable to fermionic wave functions. This also means that both the electrons occupy both the states simultaneously and hence their properties (resulting from the electronic states which they occupy) become identical as typically expected for indistinguishable particles. However if state A corresponds to a localized state and state B corresponds to an itinerant state and (assuming electron#1 to be localized and electron#2 to be itinerant) then we put electron#1 in state A and electron#2 in state B then the resulting two electron state must be represented by $|1\rangle_A|2\rangle_B$ instead of $|1\rangle_A|2\rangle_B - |2\rangle_A|1\rangle_B$ since in the latter state electron#1 is both localized and itinerant simultaneously (similar conclusion holds for electron#2 also) which is inconsistent with fact that electron#1 is localized (and similar conclusions should be drawn for electron#2). Thus incorporating fermionic exchange symmetry into such a two electron state leads to unreasonable conclusions and hence we must conclude here that the above two electron state cannot have the exchange symmetry typical of fermions (i.e. indistinguishable particles) in such cases. In essence we see that a two electron state corresponding to a localized and an itinerant electron cannot have the exchange symmetry typical of fermions. In analogy, the high temperature state of the single ion Kondo model/Kondo lattice model wherein 4*f* electrons get localized must also not possess the fermionic exchange symmetry as is seen for the low temperature state (e.g. $|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v$) and hence the localized 4*f* electrons at high temperatures must not be regarded as fermions. Instead the high temperature state must be written as $|\uparrow\rangle_f|\downarrow\rangle_v$ (or $|\downarrow\rangle_f|\uparrow\rangle_v$) which is devoid of the fermionic exchange symmetry.

B. Origin of Non-Fermi Liquid Behavior in Heavy Fermion Systems - Another Explanation

Landau's Fermi liquid theory claims that every many-electron system will possess fermionic quasiparticles as its low energy excitations no matter however large is the strength of electron correlations amongst them. Hence every electronic system will have a Fermi liquid ground state according to Landau. However, recent observations of non-Fermi liquid (NFL) behavior among many correlated electron systems have generated intense enquiry towards sorting out its origin.

In this manuscript, we argue that Landau overlooked a fundamental issue in quantum mechanics that an electronic system must possess identical spatial profile of wave function of every constituent electron in order to model the electronic system within the Fermi liquid theory. Such an identical profile ensures 'indistinguishability' amongst all the constituent electrons which is a necessary condition for them to be treated as 'fermions' obeying Fermi liquid theory. Such 'indistinguishability' can be realized in itinerant electron systems. These 'fermions' then give rise to the Fermi liquid ground state, regardless of the strength of correlations (e.g. as in paramagnetic heavy fermion systems). ***In the context of condensed matter systems, this means that the Fermi liquid theory is applicable for the itinerant 'valence' electrons but not for the localized electrons. In other words, itinerant 'valence' electrons are 'fermions'. Localized electrons are not 'fermions'***.

In the context of Kondo lattices, this idea gives rise to remarkable consequences: It is known that the ground state of a Kondo lattice possessing stable local magnetic moments is long range magnetically ordered. Here, only the itinerant valence electrons are fermions which alone constitute the valence band giving rise to Fermi liquid behavior. Localized 'magnetic' electrons are not fermions and do not participate in the valence band. On the other hand the Kondo screened ground state of a Kondo lattice is paramagnetic with all the localized 'magnetic' electrons becoming itinerant due to their quantum entanglement with the itinerant valence electrons. In this case, both the itinerant valence electrons and 'magnetic' electrons are fermions participating in the valence band. When we tune the ground state of a Kondo lattice towards the quantum critical point (QCP) from the magnetically ordered state (via variation of a control parameter like chemical/physical pressure, magnetic field etc.), we gradually introduce itinerancy in the localized 'magnetic' electrons. Consequently they start (after progressively moving closer to the Fermi energy) contributing to the valence band and affecting low energy physical properties of compounds close to QCP. However we claim that at QCP the Kondo screening process is not yet complete. Hence the 'magnetic' electrons possess partial localized character even at QCP which disqualifies them to be called as fermions. Consequently the valence band (containing both itinerant 'valence' electrons and partially localized 'magnetic' electrons) cannot be modeled within Fermi liquid theory and hence NFL behavior emerges at QCP.

C. Temperature Dependence of the Kondo Coupling

The Hamiltonian for an impurity local magnetic moment $\vec{S}$ coupled to the conduction electron $\vec{s}$ within the Kondo model is:

$$H = \sum_{k\sigma} \varepsilon_k c^{\dagger}_{k\sigma} c_{k\sigma} - J\vec{S}.\vec{s}$$

where $\vec{s} = \frac{1}{N}\sum_{k,k',\sigma,\sigma'} c^{\dagger}_{k\sigma}\sigma_{\sigma,\sigma'}c_{k',\sigma'}$ is the conduction electron spin at the impurity site (σ are the Pauli matrices; N is the number of sites)

$\varepsilon_k \rightarrow$ Conduction electron dispersion

$c^{\dagger}_{k\sigma}, c_{k\sigma} \rightarrow$ Creation and annihilation operator for an electron with wave vector *k* and spin σ, respectively

J → Kondo exchange coupling constant ($J < 0$ for antiferromagnetic Kondo coupling between the local moment and the conduction electron spin)

{Note: Although the Kondo effect is known to be a many-body problem involving many electrons (~10^{22} electrons/cm^3 - typical electron densities in metals) we attempt to illustrate the details about the Kondo coupling using an example of a two electron system (consisting of a localized magnetic electron and a conduction electron) under the justification that such details can be studied qualitatively using the two electron system. The effect of remaining electrons will only (at maximum) be to renormalize the value of the Kondo coupling J and will not change the qualitative characteristics of J like, e.g. its temperature dependence. **As a matter of fact an exchange among electrons is by definition between two electrons**. Hence our use of such two electron system is well justified and is very helpful for illustrational simplicity.}

The prevalent view within the scientific community assumes J to be constant with temperature. However, we argue that the value of J increases with decreasing temperature corresponding to the increase in the Kondo coupling strength at lower temperatures. This can be understood qualitatively in the following way. It should be noted that J is proportional to the exchange integral (J_{ex}) between the two electrons which is a function of their wave functions' spatial distribution as well as their mutual spatial separation (ref. http://en.wikipedia.org/wiki/Exchange_interaction). Specifically J_{ex} is inversely proportional to the magnitude of their mutual spatial separation. As the temperature is lowered, due to the attractive interaction between the local moment and the conduction electron, there is a tendency for both of them to stay close to each other in space thus reducing their mutual spatial separation and thereby increasing the exchange integral J_{ex} between the two electrons. Consequently the value of the Kondo exchange coupling J increases too, at lower temperatures. According to the current belief all the temperature dependent phenomenon in Kondo systems is a result of the variation of the relative strengths of the energy scales related to the thermal fluctuations to that of various exchange energy scales (e.g. Kondo scale, RKKY scale etc.) inside the solid. However, we argue that the temperature causes changes in the value of J affecting the properties of Kondo systems in addition to the usual effects caused by the thermal fluctuations.

We argue that $J \to 0$ as $T \to \infty$ and $J \to -\infty$[1] as $T \to 0$ ($J < 0$ always for antiferromagnetic Kondo coupling). $J(T)$ is a monotonic function of temperature.

*1) **Exchange Symmetry and Fermionic Behavior***

Assume the two electron system to represent a localized magnetic 4*f* electron interacting with an itinerant valence electron by the Kondo interaction. At 0 K the system forms a stable non-magnetic Kondo singlet state denoted by $|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v$. This state actually corresponds to $J \to -\infty$ wherein full exchange between both the electrons is realized. As a result we have full indistinguishability between them (their wave function being antisymmetric with respect to a particle exchange). As the magnitude of J decreases with increasing temperature, the exchange between both the electrons becomes weaker concomitantly. As a result we do not have full exchange between both the electrons instead only a partial exchange is realized between the two at non-zero finite temperatures. Consequently they are not fully indistinguishable between themselves but instead they should be considered as partially indistinguishable (what we mean by a partial exchange between the two electrons will be clear in the subsequent discussions). Thus we have a scenario in which the exchange between the two electrons is temperature dependent. The $J \to 0$ state at $T \to \infty$ corresponds to the local moment state in which both the electrons become completely distinguishable. The question is how to express the two electron state at intermediate temperatures? We claim that an arbitrary temperature state could in general be written as: $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$. {It is important to note here that in the expression $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$, $|\uparrow\rangle_f$ and $|\uparrow\rangle_v$ denotes 'total' wavefunctions for the 4*f* and valence electrons respectively i.e. spatial and spin part of their wave functions together and not just the spin parts of their wave functions as it might misleadingly look like}, wherein the second term in the expression has acquired a phase factor different from that of the first term. Thus the weight of the second term which exchanges with the first term has reduced by a factor of $\cos\theta$ as against the Kondo singlet state in which the second term exchanges with the first term completely. Such a situation amounts to a partial exchange between both the electrons giving rise to partial indistinguishability between them. Such a scenario cannot be captured within the (present-day version of) quantum mechanical formalism and requires additional principles to be introduced into it while handling such cases. Note that θ should be expressed as $\theta = (\pi/2)*\alpha$ where $0 \le \alpha \le 1$ and α depends on temperature via $J(T)$. Thus $\alpha = \alpha(J(T))$ and $\alpha \to 0$ as $T \to 0$ and $\alpha \to 1$ as $T \to \infty$. $\alpha = 0$ corresponds to the Kondo singlet state while $\alpha = 1$ is the high temperature state where full local moment is obtained and which is devoid of antisymmetry (with respect to a particle exchange). Note that α should be monotonic function of temperature.

The fermionic behavior that we encounter in physics is a result of indistinguishability of the electrons. At any moment the Fermi surface (FS) volume of an electronic system will be proportional to the weight of the part of its wave function which obeys the antisymmetry (corresponding to indistinguishable electrons). Thus for the Kondo singlet state $|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v$, due to its full indistinguishability, we must count the *f*-electron wholly inside the FS. Thus the FS contains the valence electron (by default) and also the *f*-electron wholly {Please note that in the context of Kondo systems, the valence electrons are by default considered to be itinerant and participating in the FS volume. The open issues in Kondo systems concern the participation of the localized 4*f* electron within the FS volume with varying temperature and/or non-thermal 'control' parameter}. The FS volume corresponds to

[1] $J \to -\infty$ is only an approximation as the zero point motion of the electron in quantum theory does not allow J to blow up at 0 K. Instead J remains finitely large at 0 K.

two electrons in that case. Now let us take an arbitrary temperature state i.e. $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$. We have already argued that this state does not correspond to full indistinguishability between both the electrons due to the absence of full antisymmetry (with respect to a particle exchange) for this state. However when we rewrite the above state as $(1-\cos\theta)|\uparrow\rangle_f|\downarrow\rangle_v + \cos\theta(|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v) - i\sin\theta|\downarrow\rangle_f|\uparrow\rangle_v$, , one can see that a part of this state (i.e. the middle term) obeys the antisymmetry while the rest (i.e. the remaining terms) does not. It is here we claim that the state $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$ actually manifests *partial* indistinguishability (due to the existence of the exchange between a part of the 4*f* electron, i.e. cosθ fraction of the 4*f* electron, with the valence electron) between the two electrons. Only the part of the wave function which obeys the antisymmetry contributes to the FS volume while the rest does not contribute so and therefore it corresponds to the distinguishable electronic weight→ non-fermions {Please note that the distinguishability/indistinguishability of an electron is always defined in relation to some other electron. It is not obviously clear how this will affect the FS volume. Here we argue that typically in Kondo systems the valence electrons are considered as 'benchmark' for the electrons contributing to the FS volume. Therefore any other electron (e.g. 4*f* electron in Kondo systems) becoming indistinguishable with the valence electron must be counted in the FS volume and vice versa. The intermediate case of partial indistinguishability between them would naturally lead to a partial contribution of the 4*f* electron to the FS volume}. Thus in this case the total FS volume would contain the valence electron (by default) and cosθ fraction of one *f*-electron. Then the FS volume will be (1+cosθ). Thus one can see that with increasing temperature the FS volume reduces from 2 (at T = 0K) to 1 (at T = ∞K) following the expression (1+cosθ) = (1+cos((π/2)*α) which depends on the temperature through *J*(T). The exact functional form of *J*(T) is unknown and is an open and complex issue.

This was the illustration done for a simple two electron system highlighting the intricacies of the two electron exchange as a function of temperature. Of course for real many-electron systems there might arise additional degrees of complexity and the exact FS evolution with temperature might need certain renormalization of the parameters presented here. However, the basic idea presented in this section (regarding the two electron exchange) needs to be utilized in any realistic theory simulating the temperature dependent FS evolution in Kondo lattices.

2) ***Non-Fermi Liquid (NFL) Behavior at the Quantum Critical Point (QCP)***

Instead of varying the temperature for tuning the value of *J*, one can also tune it by changing a non-thermal 'control' parameter (ρ) like physical/chemical pressure, applied magnetic field etc. Such a control parameter tuning has led to the discovery of QCP's in the heavy fermion systems which are a very active field of research currently. An interesting observation of NFL behavior close to QCP's has turned into a big open problem in contemporary condensed matter research.

We argue that our treatment about the state of the two electron system as described above is maintained even in the present case when we tune the ground state of the heavy fermion system by varying ρ. Along the ρ axis, (in case of the two electron system involving an interaction of the 4*f* electron with an effective field simulating the effect of other 4*f* sites of the Kondo lattice on the 4*f* electron → simulating the RKKY interaction (this is an approximation for the real situation in a Kondo lattice but it is quite helpful for highlighting our ideas over the variation of the two electron exchange with ρ)) if ρ→∞ corresponds to $J(\rho)\rightarrow -\infty$ (Kondo singlet ground state, $|\uparrow\rangle_f|\downarrow\rangle_v - |\downarrow\rangle_f|\uparrow\rangle_v$) and ρ→0 corresponds to $J(\rho)\rightarrow 0$ (local magnetic moment ground state) then any ground state for a finite ρ will in general be represented by $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$, where θ = (π/2)*α; 0≤α≤1 and α depends on ρ via *J*(ρ). Close to the critical value of ρ, i.e. ρ_C where the QCP is believed to exist, the induced itinerancy into the *f*-electron (by increasing the magnitude of *J*(ρ)) is large enough for it to influence the physical properties of the systems although $\rho_C<\infty$. Thus the *f*-electron becomes a part of the valence band although it has not fully matured into a fermion yet. This leads the valence band to display NFL excitations.

In case of a Kondo lattice, the real situation is far more complex due to the RKKY interaction but the basic details about the two electron exchange presented here must be utilized for a realistic simulation of the variation of the ground state of heavy fermion systems with ρ. This holds as well for the study of the variation of the FS volume with ρ, in which case we admit that a large amount of conflicting experimental evidence is available as far as the exact location of large-small FS transition with respect to the QCP is concerned. However our approach of treating the two electron exchange in a novel way, we believe, would allow a large degree of flexibility for accommodating a wide variety of ground states within its predictions.

3) ***On the Origin of the Temperature Dependence of the Kondo Coupling***

Assuming a two electron state at any arbitrary temperature as $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$, we can show (from ref. http://en.wikipedia.org/wiki/Exchange_interaction) that the Kondo exchange coupling *J* is temperature dependent. We proceed as follows:

The Kondo Hamiltonian involves a Heisenberg exchange term containing coupling of the local moment spin with the conduction electron spin i.e. a term of the form $-J\vec{s}_a.\vec{s}_b$. Now $\vec{s}_a.\vec{s}_b$ is known to have characteristic values corresponding to singlet ($\vec{s}_a.\vec{s}_b=-3/4$) and triplet ($\vec{s}_a.\vec{s}_b=1/4$) spin wave functions for the two electron system as illustrated in the aforementioned internet reference. Since we wish to estimate the value of *J* for any arbitrary temperature, we need to assume the two electron wave function to be given by the form $|\uparrow\rangle_f|\downarrow\rangle_v - e^{i\theta}|\downarrow\rangle_f|\uparrow\rangle_v$ which has an intermediate exchange symmetry (i.e. between bosonic and fermionic exchange symmetry) as against the antisymmetry which is typical of fermions. Hence we need to construct the electron wave functions from their spatial and spin parts in such a way that the 'total' electron wave functions possess such an intermediate symmetry. For illustrating this concept we use the case of the H_2 molecule as discussed in the aforementioned

internet reference. Since the total spin of the two electron system is a good quantum number (for both the Hamiltonians i.e. for H_2 molecule and the Kondo Hamiltonian) the eigenstates of the two electron Hamiltonian have to be constructed in such a way that the intermediate exchange symmetry of their wave functions is contained in their spatial part but not the spin part. This construction scheme guarantees the existence of intermediate exchange symmetry for the 'total' two electron wave function while keeping their spin parts as eigenstates of the operator corresponding to the total spin of the two electron system. Let the eigenstates of the single electron Hamiltonian for the individual H atom (as in the Heitler-London approximation) be labeled by ψ_a^0, ψ_a^1, ψ_a^2, ψ_a^3,….. for atom *a* and ψ_b^0, ψ_b^1, ψ_b^2, ψ_b^3,….. for atom *b* where ψ_a^0 and ψ_b^0 represent their ground states respectively, we argue that at temperature T the electronic state for atom *a* and atom *b* should be written as:

$$\psi_a(\vec{r},\sigma,T)=\sum_{i=0}^{\infty}c_a^i(T)\psi_a^i(\vec{r})=\sum_{i=0}^{\infty}c_a^i(T)\phi_a^i(\vec{r})\chi_a^i(\sigma)=\sum_{i=0}^{\infty}\tilde{\phi}_a^i(\vec{r},T)\chi_a^i(\sigma)\ \text{(for atom a)}$$

and $\psi_b(\vec{r},\sigma,T)=\sum_{i=0}^{\infty}c_b^i(T)\psi_b^i(\vec{r})=\sum_{i=0}^{\infty}c_b^i(T)\phi_b^i(\vec{r})\chi_b^i(\sigma)=\sum_{i=0}^{\infty}\tilde{\phi}_b^i(\vec{r},T)\chi_b^i(\sigma)$ (for atom *b*)

where ϕ_a^i,χ_a^i are spatial and spin parts of the single electron wavefunction for atom *a* respectively and ϕ_b^i,χ_b^i are spatial and spin parts of the single electron wavefunction for atom *b* respectively and $c^i_a(T)$, $c^i_b(T)$ are the temperature dependent occupancies of the eigenstates of the Hamiltonian for atom *a* and atom *b* respectively. Note that the wavefunctions $\{\psi_a^i\},\{\phi_a^i\},\{\chi_a^i\}$ are normalized and orthogonal to each other within their respective families (denoted by curly brackets) i.e. $\langle\psi_a^i|\psi_a^j\rangle=\delta_{ij};\langle\phi_a^i|\phi_a^j\rangle=\delta_{ij};\langle\chi_a^i|\chi_a^j\rangle=\delta_{ij}$. Similar is the case with $\{\psi_b^i\},\{\phi_b^i\},\{\chi_b^i\}$.

In order to illustrate the temperature dependence of *J* it is convenient to describe the wavefunctions in terms of molecular orbitals. We construct molecular orbitals (M.O.) for the H_2 molecule as:

$U_p(\vec{r},T)=N_p\sum_{i=0}^{\infty}\frac{\tilde{\phi}_a^i(\vec{r},T)+\tilde{\phi}_b^i(\vec{r},T)}{\sqrt{2(1+\alpha_i)}}$ and $U_q(\vec{r},T)=N_q\sum_{i=0}^{\infty}\frac{\tilde{\phi}_a^i(\vec{r},T)-\tilde{\phi}_b^i(\vec{r},T)}{\sqrt{2(1-\alpha_i)}}$

where $\alpha_i=\int\tilde{\phi}_a^{i^*}(\vec{r},T)\tilde{\phi}_b^i(\vec{r},T)\,d^3r$ is the overlap integral and N_p & N_q are normalizing factors.

Note that $U_p(\vec{r},T)$ and $U_q(\vec{r},T)$ are orthogonal to each other.

Then the symmetric and anti-symmetric combinations from them can be created as:

$\psi_A(\vec{r}_1,\vec{r}_2,T)=\frac{1}{\sqrt{2}}\left[U_p(\vec{r}_1,T)U_q(\vec{r}_2,T)-U_q(\vec{r}_1,T)U_p(\vec{r}_2,T)\right]$

and $\psi_S(\vec{r}_1,\vec{r}_2,T)=\frac{1}{\sqrt{2}}\left[U_p(\vec{r}_1,T)U_q(\vec{r}_2,T)+U_q(\vec{r}_1,T)U_p(\vec{r}_2,T)\right]$

However in order to acquire intermediate exchange symmetry at finite temperatures we must write the above wave functions (including spin part) as:

$\Psi_A(\vec{r}_1,\vec{r}_2;\sigma_1,\sigma_2,T)=\frac{1}{\sqrt{2}}\left[U_p(\vec{r}_1,T)U_q(\vec{r}_2,T)-e^{i\theta}U_q(\vec{r}_1,T)U_p(\vec{r}_2,T)\right]\otimes\{\text{Normalized spin part of the spin triplet wave function}\}$ And

$\Psi_S(\vec{r}_1,\vec{r}_2;\sigma_1,\sigma_2,T)=\frac{1}{\sqrt{2}}\left[U_p(\vec{r}_1,T)U_q(\vec{r}_2,T)+e^{i\theta}U_q(\vec{r}_1,T)U_p(\vec{r}_2,T)\right]\otimes\{\text{Normalized spin part of the spin singlet wave function}\}$

Breaking the Hamiltonian into two parts as $H=H^0+H^1$ (as in M.O. approximation) where H^0 is the unperturbed Hamiltonian and $H^1\propto\frac{1}{r_{12}}$ is the perturbation comprising of inter-electron interactions. Note that H does not contain any spin operators and hence it will not operate on the spin part of the wave functions. Then the energy eigenvalues after making the first order correction to them within the perturbation theory come out to be;

$E_S=E^0+\langle\Psi_S|H^1|\Psi_S\rangle=E^0+C(T)+\cos\theta.J_{ex}(T)$

and

$E_A=E^0+\langle\Psi_A|H^1|\Psi_A\rangle=E^0+C(T)-\cos\theta.J_{ex}(T)$

where *C* is the Coulomb integral and J_{ex} is the exchange integral defined as

$C(T)=\int U_p(\vec{r}_1,T)^2H^1U_q(\vec{r}_2,T)^2d^3r_1d^3r_2$

and $J_{ex}(T)=\int U_p(\vec{r}_1,T)U_q(\vec{r}_2,T)H^1U_q(\vec{r}_1,T)U_p(\vec{r}_2,T)d^3r_1d^3r_2$

Then $J(T)=E_S(T)-E_A(T)=2\cos\theta.J_{ex}(T)$

where $\theta(T)$ and $J_{ex}(T)$ depends on temperature. Hence *J* depends on temperature. The temperature dependence of $J_{ex}(T)$ is due to the temperature dependence of the wavefunctions' spatial profile and the inter-electron spatial separation. At high temperatures $\psi_a(\vec{r},\sigma,T)$ and $\psi_b(\vec{r},\sigma,T)$ contain contributions from excited states which have extended wavefunctions in space which would naturally increase inter-electron separation. Since the exchange interaction originates from the Coulomb interaction between the electrons it is bound to become stronger when the electrons are close together than when they are far apart which happens when we reduce temperature. Then J_{ex} must increase at low temperatures. This forms a justification for claiming an intrinsic origin for

the temperature dependence of *J* in addition to usual thermal fluctuation effects affecting the physics of Kondo systems. Kindly note the importance of $e^{i\theta}$ factor in these expressions. Without this factor we are unable to find explanation for non-Fermi liquid behavior observed close to QCP. In addition, this factor (together with the temperature dependence of $J_{ex}(T)$) may be a crucial component in the glue for Cooper pairs in unconventional superconductors which needs to be explored further.

Note that it is a convention in the condensed matter community to describe the high temperature electronic state of any system as a 'mixed' quantum state rather than a pure quantum state which has been used in this manuscript all throughout for illustrating our new ideas. Such a mixed state is dealt within the density matrix formalism. The ideas presented by us should be extended to the mixed states within the density matrix formalism for a proper simulation of condensed matter phenomena.

4) ***Final Discussion***

A lot of experimental investigation has been done in the past demonstrating the existence of Kondo effect at an atomic level (e.g. Science **281**, 540 (1998), PRL **80**, 2893 (1998), Science **280**, 567 (1998) etc.). A modification of those experiments for the purpose of measuring the force between a localized spin and the conduction electrons is required for studying the temperature dependence of *J* and thereby verifying its temperature dependence experimentally. This can be achieved in one way by measuring the small attractive force produced on a single magnetic adatom deposited on a non-magnetic metallic surface, as a function of temperature. However, it is important to ensure here that the adatom electronic states hybridize with the conduction electron states in order to give rise to the Kondo effect and thereby to the existence of Kondo exchange coupling *J*. In the example of Co adatoms on Au(111) surface, this system is well known to display such a Kondo effect and has also been used previously to verify the spectroscopic signature of Kondo resonance at an atomic level for the first time (ref. Science **280**, 567 (1998)). Further, it is also important to ensure that the hybridization strength between adatom states and conduction electron states remain constant with temperature while measuring the force on the adatom. For the Co/Au(111) system as discussed above the hybridization depends on the overlap of the Co *d*-states with the conduction electron states which is dependent on the distance of the Co adatom from the Au(111) surface. This distance could change with temperature however since the thermal contraction effects etc. on the lattice constants of solids are usually very small on a relative scale it is likely that in this case to the changes in the Co-Au(111) interatomic distance would not be drastic. However such a change, if any, needs to be measured and corrected. We believe that the experiments performed on Co/Au(111) system are most likely to be successful in verifying the temperature dependence of *J*. Another example of such an experiment would be to measure the 'force' between a quantum dot (kept under a fixed gate voltage fixing the strength of the so-called hybridization between localized spin and the conduction electron reservoir) and source/drain leads (the experimental assembly as mentioned in ref. Science **281**, 540 (1998)) as a function of temperature one 'may be' able to study the temperature dependence of *J*. In the latter experiment there seems to be a large degree of control over fixing the strength of the (constant) hybridization during the experiment by fixing the gate voltage accordingly which is an adjustable parameter within the experiment as compared to the first experiment where the hybridization which depends on the distance between the Co adatom and the Au(111) metal surface seems not readily controllable in the experiment.

Our manuscript is mainly aimed at providing a crucial conceptual input at the most fundamental level which has been overlooked so far in various theoretical models simulating diverse phenomena in condensed matter instead of providing a complete theoretical formalism for simulating exact details of various condensed matter systems for a direct comparison with the experiment. Nevertheless the impact of our conceptual input seems very wide covering areas dealing with the temperature dependent transformation of electronic character in correlated electron systems, various competing ground states in condensed matter, quantum criticality and emergent non-Fermi liquid behavior and remarkably a potential candidate for a glue for unconventional superconductivity. All these big problems in contemporary condensed matter physics seem connected at the very basic level through this conceptual input. Furthermore we do admit that although DMFT has had good enough success for capturing the temperature dependent localization-itinerant transition in some of the heavy fermion compounds and currently enjoys the reputation of being the most powerful tool for simulation of correlated electron phenomena in condensed matter systems however our conceptual picture has the advantage of interweaving different condensed matter phenomena into one fabric. Therefore we believe that our manuscript does represent a significant advance in science which would make remarkable impact on future theoretical models in condensed matter physics.