RKKY Interaction and Intrinsic Frustration in Non-Fermi-Liquid Metals

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We study the RKKY interaction in non-Fermi-liquid metals. We find that the RKKY interaction mediated by some non-Fermi-liquid metals can be of much longer range than for a Fermi liquid. The oscillatory nature of the RKKY interaction thus becomes more important in such non-Fermi liquids, and gives rise to enhanced frustration when the spins form a lattice. Frustration suppresses the magnetic ordering temperature of the lattice spin system. Furthermore, we find that the spin system with a longer range RKKY interaction can be described by the Brazovskii model, where the ordering wave vector lies on a higher dimensional manifold. Strong fluctuations in such a model lead to a first-order phase transition and/or glassy phase. This may explain some recent experiments where glassy behavior was observed in stoichiometric heavy fermion material close to a ferromagnetic quantum critical point.

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Introduction.—When magnetic moments are placed in a metal, the conduction electrons mediate an indirect interaction between these moments. Such a long rang interaction is called the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. RKKY interaction plays crucial roles in, e.g., heavy fermions, diluted magnetic semiconductors, graphene. The usual derivation of the RKKY interaction is based on the assumption that the conduction electrons form a Landau Fermi liquid (FL). However many strongly correlated electron systems show non-Fermi liquid (NFL) behavior, e.g., cuprates, heavy fermions, pnictides. The question we ask here is what is the form of RKKY interaction in a NFL metal, and what are the consequencies.

Of particular interest are heavy fermion systems, where local moments couple to the conduction electrons. The Doniach phase diagram with competing Kondo coupling and RKKY interaction has been the paradigm for heavy fermions for decades [1]. In the last few years, as experimental results accumulate, there is a growing necessity to go beyond the Doniach phase diagram. Frustration or the quantum zero point energy has been proposed as a new dimension in the global phase diagram of heavy fermions [2–6]. One obvious origin of frustration is frustration of lattice structure itself. However such geometric frustration is not universally observed in heavy fermion materials. Here we propose that the NFL nature of conduction electrons in the Kondo liquid phase leads to intrinsic frustration for the localized spin degrees of freedom. This provides a more universal source of frustration.

Our approach is based on the idea of quantum criticality and NFL behavior. The standard picture is that the critical fluctuations near a quantum critical point (QCP) lead to NFL behavior. Here we depart from this picture by starting with the assumption that in a certain range of the parameter space, the itinerant electrons form a NFL state. We then proceed to study its consequences on other degrees of freedom, e.g., the localized spins. Focusing on the regime with small Kondo coupling, i.e., a small Fermi surface, we find that the magnetic transition temperature will be reduced by the frustration resulting from longer-range RKKY interaction produced by NFL itinerant electrons. Furthermore, we find that the putative ferromagnetic (FM) QCP may be replaced by a first-order phase transition or a glassy phase [7,8] (see Fig. 1).

Formalism.—We start with the Kondo lattice model, $H = H_C + H_K$. Here H_C is the conduction electron Hamiltonian, and ususally only the hopping term is included. The Kondo coupling between conduction electrons and localized spins is of the form, $H_K = -(J_K/2)\sum_{i\alpha\beta}\boldsymbol{S}_i \cdot c^\dagger_{i\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{i\beta}$. We depart from the usual approach by considering the conduction electrons to be strongly interacting themselves, i.e., $H_C = H_C^{(0)} + H_C^{(\mathrm{int})}$. One way to motivate this is to consider the phenomenological two fluid model [9–12]. In many heavy fermion systems, below the coherence temperature T^*, the experimental results can be understood in terms of the two fluid model, with one component the itinerant heavy electrons, and the other component local moments. The heavy electron Kondo liquid is not a simple FL; e.g., the specific heat is logarithmically enhanced at low temperature. One has a model of interacting itinerant electrons coupled with localized spins.

The itinerant electrons induce a RKKY type interaction among the localized moments,

$$H_{\mathrm{RKKY}} = \sum_{ij} J_{ij}^{ab} S_i^a S_j^b. \tag{1}$$

Here the coupling $J_{ij}^{ab} = -(J_K^2/4)\chi_{ij}^{ab}$ [13–15], is determined by the static spin susceptibility of the conduction electrons

$$\chi_{ij}^{ab} = -\frac{i}{\hbar}\int_0^\infty \langle[s^a(\boldsymbol{r}_i, t), s^b(\boldsymbol{r}_j, 0)]\rangle e^{-\eta t} dt, \tag{2}$$

with the electron spin $s^a(\boldsymbol{r}_i) = \sum_{\alpha\beta} c^\dagger_{i\alpha}\sigma^a_{\alpha\beta}c_{i\beta}$ and $\eta = 0^+$. If the conduction electrons are in the paramagnetic state,

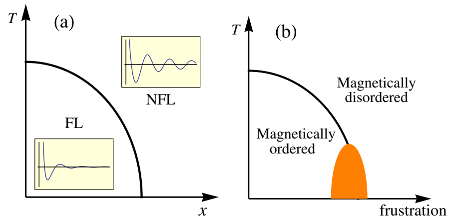

FIG. 1 (color online). Schematic electronic (a) and magnetic (b) phase diagrams for a Kondo lattice model with strongly interacting conduction electrons coupled to localized spins. (a): Crossover from FL to NFL behavior. x represents non-thermal tuning parameter, e.g., pressure, magnetic field, doping. Distance dependence of RKKY interaction is shown in the insets. (b): Magnetic transition temperature decreases with increasing frustration resulting from NFL-mediated longer-range RKKY interaction. New phases (shaded region), e.g., a glass phase, emerge near the putative QCP.

the spin susceptibility is isotropic and diagonal, i.e., $\chi_{ij}^{ab} = \chi(\boldsymbol{r}_{ij})\delta^{ab}$. For FL, the spin susceptibility behaves as $\chi(\boldsymbol{r}) \sim (1/r^d)\cos(2k_F r + \theta_0)$ at long distances, with d the spatial dimension. This leads directly to the standard form of the RKKY interaction. The exponent d results from the sharp jump in the momentum distribution $n(\boldsymbol{k})$, characteristic of FL. For NFL metals, the RKKY interaction can have qualitatively different behavior. We still assume the existence of a Fermi surface, i.e., a singularity in $n(\boldsymbol{k})$; thus, the spin susceptibility still has $2k_F$ oscillation. The exponent can take a different value. Thus we have $\chi(\boldsymbol{r}) \sim (1/r^\alpha) \times \cos(2k_F r + \theta_0)$. More detailed studies of the NFL spin susceptibility will be presented below.

Consider placing a lattice of spins in the NFL metal. We focus on the effect of the RKKY interaction on the spin system, and will not consider the competition between Kondo coupling and RKKY interaction [1]. This can be achieved by assuming the spins to be classical, or considering only the part of the phase diagram with a small Fermi surface. With $\boldsymbol{S}(\boldsymbol{q}) = (1/N)\sum_i \boldsymbol{S}_i e^{i\boldsymbol{q}\cdot\boldsymbol{r}_i}$, one has in momentum space, $H = \sum_{\boldsymbol{q}} F(\boldsymbol{q})\boldsymbol{S}(\boldsymbol{q})\cdot\boldsymbol{S}(-\boldsymbol{q})$, where

$$F(\boldsymbol{q}) = \frac{1}{N}\sum_{\boldsymbol{r}_i \neq 0} J(\boldsymbol{r}_i) e^{i\boldsymbol{q}\cdot\boldsymbol{r}_i}, \tag{3}$$

with $\boldsymbol{r}_i$ defined on the lattice. The ordering wave vector in the ground state is determined by minimizing the function $F(\boldsymbol{q})$.

For the conventional three dimensional RKKY interaction mediated by FL, this problem has been studied in [16], where different phases have been identified as the conduction electron density changes. At small $k_F a$, where a is the lattice constant, the ground state is ferromagnetic. As $k_F a$ increases, antiferromagnetic phases with different ordering wave vectors appear. In the case $k_F a \to 0$, the above summation can be replaced by an integral, and $F(\boldsymbol{q}) \sim -\chi(q)$. The ordering wave vector is thus determined by maximizing the static spin susceptibility.

Now we consider in more detail what is the form of the static spin susceptibility in a NFL metal. When vertex corrections can be ignored, the spin susceptibility can be calculated from the fermion bubble, with $\chi_{ab}(q) \sim \int \sigma^a G(k+q)\sigma^b G(k)$. When the momentum distribution $n(\boldsymbol{k})$ has a weaker singularity than a jump at k_F, e.g., a kink, the single particle density matrix $n(\boldsymbol{r})$ decays faster than that of FL (see Supplemental Material [17]). Then one expects $\chi(r)$ and $J(r)$ to decay faster than that of FL. An interesting question is whether it is possible to have longer range RKKY interactions, which would generate the desired frustration among the spins [2–6]. We will present two models of NFL metals that can give rise to such behavior.

Longer range RKKY interaction in one dimension.—First, as a proof of principle that RKKY interaction in a strongly interacting electron system can be of longer range than in a free system, let us first consider one dimension. In one dimension, RKKY interaction mediated by free electrons is of the form $J(r) \sim \mathrm{Si}(2k_F r) - \pi/2$, with the sine integral function $\mathrm{Si}(x)$. At large distance one has $J(r) \sim \cos(2k_F r)/r$. In momentum space, one has $\chi(q) \sim (1/q) \times \ln|(q + 2k_F)/(q - 2k_F)|$, with a maximum at $q = 2k_F$.

The low energy dynamics of interacting electrons in one dimension is described by the Luttinger liquid theory. Due to spin-charge separation, the conduction electron Hamiltonain can be written as a summation of the two channels [18],

$$H_C = \sum_{\alpha=c,s} \frac{v_\alpha}{2} \int dx[g_\alpha \Pi_\alpha^2 + g_\alpha^{-1}(\partial_x\theta_\alpha)^2], \tag{4}$$

with v_c and v_s the velocity of charge and spin density wave, respectively. The charge interaction constant $g_c = 1$ for noninteracting fermions, $g_c < 1$ for repulsive interaction, and $g_c > 1$ for attractive interaction. We are interested in the case with repulsive interaction. The spin interaction constant $g_s = 1$ in the presence of SU(2) spin symmetry. The oscillating part of the spin correlation function is [18]

$$\langle \boldsymbol{s}(x,\tau)\cdot\boldsymbol{s}(0,0)\rangle \sim \frac{\cos(2k_F x)}{|\tau + ix/v_c|^{g_c}|\tau + ix/v_s|^{g_s}}. \tag{5}$$

The RKKY interaction, determined from the static spin susceptibility, is of the form

$$J(x) \sim \int d\tau \frac{\cos(2k_F x)}{|\tau + ix/v_c|^{g_c}|\tau + ix/v_s|^{g_s}} \sim \frac{\cos(2k_F x)}{x^{g_c+g_s-1}}. \tag{6}$$

For $g_c < 1$, $g_s = 1$, the exponent $\alpha = g_c + g_s - 1 < d = 1$. The RKKY interaction mediated by a Luttinger liquid is thus of longer range than that mediated by a non-interacting Fermi gas [19].

Spin susceptibility in two dimensions.—Now we consider two-dimensional metals. For free electrons, the static spin susceptibility reads

$$\chi(q) = \begin{cases} \chi_0 & \text{for } q < 2k_F \\ \chi_0[1 - \sqrt{1-(2k_F/q)^2}] & \text{for } q > 2k_F, \end{cases} \tag{7}$$

with $\chi_0 = 1/\pi$, which has a one-sided square-root singularity. The RKKY interaction is thus of the form $J(r) \sim \sin(2k_F r)/r^2$. For a two-dimensional FL, including higher-order diagrams, there is also a square-root singularity for $q < 2k_F$, with $\chi(q) = \chi(2k_F) + \chi^{\rm sing}(q)$ [21], and

$$\chi^{\rm sing}(q) = \mathcal{A}\sqrt{1-(q/2k_F)^2}. \tag{8}$$

The new singularity gives contribution $\delta\chi(r) \sim [(2k_F r) \times \cos(2k_F r) - \sin(2k_F r)]/r^3$, and will not change the long-distance behavior of the RKKY interaction.

A prototype of NFL metal in higher dimensions is the system of two-dimensional degenerate fermions interacting via a singular gauge interaction [22–29], where the presence of the gauge interaction leads to singular $2k_F$ response [26,30]. The fermion $2k_F$ vertex Γ_{2k_F} has a power law dependence on frequency, with $\Gamma_{2k_F} \sim (E_F/\omega)^\sigma \Gamma^0_{2k_F}$. The exponent is of the form $\sigma = 1/(2N) + 1/(2\pi^2 N^2)\ln^3 N + \mathcal{O}(1/N^2)$ for large N, and $\sigma = (16\sqrt{2})/(9\pi\sqrt{N}) + \mathcal{O}(1)$ for small N. Here the spin index is generalized to take values from 1 to N. Taking $N = 2$, one obtains $\sigma = 0.25$ from the large-N expansion, and $\sigma = 0.56$ in the small-N limit.

The spin susceptibility is calculated from the polarization bubble with vortex corrections [31], $\chi(\boldsymbol{q}, \omega) \simeq \Pi(\boldsymbol{q}, \omega) = \int d\boldsymbol{p} d\epsilon G(\boldsymbol{p} + \boldsymbol{q}/2, \epsilon + \omega/2) G(\boldsymbol{p} - \boldsymbol{q}/2, \epsilon - \omega/2) \times [\Gamma_{\epsilon\boldsymbol{p}}(\boldsymbol{q}, \omega)]^2$. For $\sigma < 1/3$, the static spin susceptibility is of the form [26]

$$\chi(q) \sim \chi_0 - \mathcal{C}|q - 2k_F|^{1-3\sigma}, \tag{9}$$

and for $\sigma > 1/3$ one has [26]

$$\chi(q) \sim \frac{1}{|q - 2k_F|^{3\sigma-1}}, \tag{10}$$

with a singularity at $q = 2k_F$ (see Fig. 2(a)). Fourier transforming to real space, we find

$$\chi(\boldsymbol{r}) \sim \int \frac{1}{|q-2k_F|^{3\sigma-1}} J_0(qr) q dq \sim \frac{\cos(2k_F r - \theta_0)}{r^{5/2-3\sigma}}. \tag{11}$$

The exponent $\alpha = 5/2 - 3\sigma$ can be much smaller than the space dimension $d = 2$.

More generally, for NFL metals, one can employ a scaling theory for the susceptibility (see, e.g., [32,33]). Assuming the existence of a Fermi surface, the static spin susceptibility generally has a power law behavior near $q = 2k_F$, with $\chi(\boldsymbol{q}) \sim |q - 2k_F|^\nu$. For $\nu < 1/2$, one has a stronger singularity than the FL case, and the RKKY interaction is of longer range.

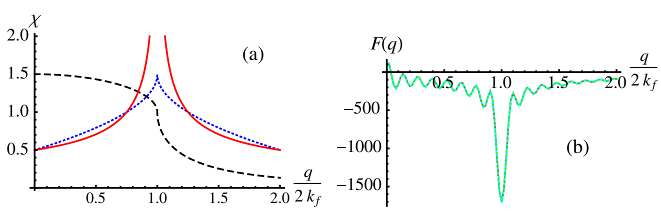

FIG. 2 (color online). (a) The static spin susceptibility $\chi(q)$ as a function of momentum for a FL (dashed, black) and the gauge-fermion model with $\sigma < 1/3$ (dotted, blue) and $\sigma > 1/3$ (solid, red). (b) $F(q)$ as a function of momentum for angles $\theta = 0$, $\pi/6$, $\pi/4$. The curves for different angles are almost identical. Here the spins form a square lattice, and $\sigma = 1/2$, $k_F a = 0.2$.

Longer range RKKY interaction in two dimensions.—Let us now consider the ground state of the spins embedded in two-dimensional metals with small $k_F a$. For the FL case [Eqs. (7) and (8)], $\chi(q)$ increases monotonically with decreasing q (see Fig. 2). The ground state is ferromagnetic. For NFL [Eqs. (9) and (10)], the maximum of $\chi(q)$ is at $q = 2k_F$, and the ferromagnetic state is no longer the ground state. More precisely, one can calculate the interaction $F(q)$ by first Fourier transforming $\chi(\boldsymbol{q})$ to real space to get $\chi(\boldsymbol{r})$, and then performing the lattice summation in Eq. (3). For simplicity we consider here the case of conduction electrons having an isotropic Fermi surface [34]. The result for $\sigma = 1/2$ is shown in Fig. 2(b). One can see that $F(q)$ has a minimum at $q = 2k_F$. The singularity in $\chi(q)$ is smeared out by the lattice effect.

Another observation is that $F(\boldsymbol{q})$ has a very weak dependence on the direction of momentum. In Fig. 2(b), $F(\boldsymbol{q})$ for the three different angles are almost indistinguishable. With the minimum of $F(\boldsymbol{q})$ at $q_0 = 2k_F$, the ordering wave vector of the lattice spin system lies on a shell of radius $2k_F$. Expanding $F(\boldsymbol{q})$ around q_0, one obtains the Brazovskii model [35],

$$H = \sum_{\boldsymbol{q}} [b_0 + D(|\boldsymbol{q}| - q_0)^2] \boldsymbol{S}(\boldsymbol{q}) \cdot \boldsymbol{S}(-\boldsymbol{q}). \tag{12}$$

Brazovskii found that the large phase space available for fluctuations around a shell of minima leads to a first-order phase transition [35]. It has been found experimentally that putative FM-QCPs are replaced by first order transitions at low temperatures in several transition metal compounds, e.g., MnSi, $ZrZn_2$, and heavy fermion systems, e.g., UGe_2, UCoAl, UCoGe (see [36] and references therein). It was realized earlier that competing orders [37] as well as fluctuations [38–41] can lead to first order quantum phase transitions. Here we find a new mechanism where the frustration resulting from NFL behavior can generate first order transitions.

A further observation is that the extensive configurational entropy in the Brazovskii model should lead to slow dynamics and glassiness [42–45]. Glassy correlations emerge when the correlation length $\xi = (D/b)^{1/2}$ becomes of order the modulation length $l_0 = 2\pi/q_0$ [43]. The parameter b needs to be determined self-consistently. Within the large-N approximation, and including a small quartic term with coupling u, we have

$$b = b_0 + uT \int \frac{d^2\boldsymbol{q}}{(2\pi)^2} \mathcal{G}(q), \tag{13}$$

with the Green's function $\mathcal{G}(\boldsymbol{q}) = 1/[b + D(q - q_0)^2]$. The condition $\xi/l_0 \sim 1$ then determines the temperature where glassy behavior sets in to be $T_g \simeq (2\pi D^2/u)(q_0^2 - b_0/D/c_1 - \log(q_0 a))$, with the coefficient c_1 of order unity and momentum cutoff $\Lambda \sim a^{-1}$. We notice that here T_g depends logarithmically on cutoff instead of the $1/\Lambda$ dependence for the three-dimensional model considered in [43].

Glassy spin dynamics was recently observed in the heavy fermion system CeFePO [46]. CeFePO is a layered Kondo lattice system, in close proximity to a FM QCP. Spin-glass-like freezing was detected in the ac susceptibility, specific heat and muon-spin relaxation [46]. The glass behavior in such a stoichiometric system points to new mechanisms that do not depend on external randomness. Our model provides such a possibility (see [47–52] and references therein for earlier attempts to obtain glass behavior from frustrated deterministic models).

Away from QCP.—We proceed to study the lattice spin system away from QCP, to see how the change of interaction range affects magnetic ordering. Due to the cosin function, the RKKY interaction changes sign and magnitude with distance. It can be well approximated by a random interaction [53–56], $J_{ij} \sim (J_K^2/4)(\epsilon_{ij}/r^\alpha)$, where ϵ_{ij} is a random variable with cosine distribution $P(\epsilon_{ij}) = (1/\pi)(1-\epsilon_{ij}^2)^{-1/2}$.

When the itinerant electrons are away from the QCP, there is a crossover to FL behavior at low energy, or equivalently long distance, where the RKKY interaction is substantially reduced. We will assume for simplicity that the RKKY interaction can be neglected beyond a crossover scale $r_{\rm FL}$. Then the exchange interaction is of the form

$$J_{ij} = \begin{cases} \mathcal{A}\epsilon_{ij}/|\mathbf{r}_i - \mathbf{r}_j|^\alpha & \text{for } |\mathbf{r}_i - \mathbf{r}_j| < r_{\rm FL} \\ 0 & \text{for } |\mathbf{r}_i - \mathbf{r}_j| > r_{\rm FL}. \end{cases} \quad (14)$$

We start with a lattice spin system that is magnetically ordered when $r_{\rm FL}$ is small. As $r_{\rm FL}$ increases, the ordering temperature will be reduced by frustration.

A simpler model that illustrates essentially the same effect of suppression of ordering by frustration is the Sherrington-Kirkpatrick model [57,58]. Consider here ferromagnetic ordering. We start with a mean field type Hamiltonian $H = -J_0 \sum_{\langle ij\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$, with $J_0 > 0$, and each spin interacting with z neighboring spins. The spins order ferromagnetically below the transition temperature $T_c^{(0)} = \tilde{J}_0 S(S+1)/6$, with $\tilde{J}_0 = zJ_0$. This corresponds to the case far away from the QCP.

Then we add to the above mean field ferromagnetic model random exchange interactions to model the frustration effect when approaching a QCP. The new Hamiltonian can be written as $H = -\sum_{\langle ij\rangle} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$, where the interaction J_{ij} is distributed according to $P(J_{ij}) = 1/\sqrt{2\pi J^2}\exp[-(J_{ij}-J_0)^2/2J^2]$ [57,58]. This model is readily solved by the replica technique [58,59], and the transition temperature to ferromagnetism is reduced by the random interactions, with the result [58,60]

$$T_c = T_c^{(0)}\left[\frac{1}{2} + \frac{1}{2}\sqrt{1 - \frac{3}{S(S+1)}\frac{\tilde{J}^2}{\tilde{J}_0^2}}\right], \quad (15)$$

where we have defined $\tilde{J} = z^{1/2}J$.

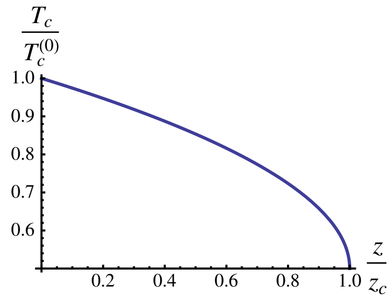

FIG. 3 (color online). Ferromagnetic transition temperature as function of range of random exchange interaction.

We fix the mean field ordering temperature in the absence of random exchange interaction $T_c^{(0)}$ and the variance of the random distribution J, so that z is a measure of the range of random exchange interaction, i.e., $r_{\rm FL}$ in Eq. (14). We can define $z_c = (S(S+1)/3)\tilde{J}_0^2/J^2$, and write T_c in the form

$$T_c = T_c^{(0)}\left[\frac{1}{2} + \frac{1}{2}\sqrt{1 - \frac{z}{z_c}}\right], \quad (16)$$

which is plotted in Fig. 3. One can see that with increasing range of random exchange interaction, the FM ordering temperature decreases. This then translates to the picture that when approaching the QCP, as RKKY interaction becomes of longer range, magnetic ordering is suppressed (see Fig. 1).

Conclusions.—We have studied RKKY interaction in NFL metals. The basic picture we find is summarized in Fig. 1. In some NFL phases, when including vertex corrections, the RKKY interaction can be of longer range than in a FL. Longer range RKKY interaction leads to frustration for the lattice spin system placed in such a NFL metal. Magnetic ordering will be suppressed by frustration, and novel behavior may emerge near the putative QCP. In particular, the continuous second-order phase transitions may be replaced by first-order transtions. Glassy dynamics may occur near the QCP without invoking disorder. One candidate material for such glassy behavior is the heavy fermion system CeFePO. We focused here on the FM QCP. One can also generalize the whole procedure to the AFM QCP by increasing $k_F a$. A further question is whether quantum fluctuations can destroy the spin glass phase and produce a spin liquid state, as in the infinite-range random-exchange model [61]. In Co- and Ge-doped $YbRh_2Si_2$, a spin-liquid-type ground state was found in the region of the phase diagram between the magnetic phase transition and Fermi-surface reconstruction [62,63]. Another interesting question is the competition between the Kondo coupling and the longer range RKKY interaction.

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