

SPIN FLUCTUATIONS AND SUPERCONDUCTIVITY AROUND THE MAGNETIC INSTABILITY*

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We summarize the present status of the theories of spin fluctuations in dealing with the anomalous or non-Fermi liquid behavior and unconventional superconductivity in strongly correlated electron systems around their magnetic instabilities or quantum critical points. Arguments are given to indicate that the spin fluctuation mechanisms is the common origin of superconductivity in heavy electron systems, 2-dimensional organic conductors and high T_c cuprates.

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1. Introduction

Anomalous physical properties and unconventional superconductivity around the magnetic instability have been the subjects of intensive current interests. Let us first look at Fig. 1, showing a phase diagram around the magnetic instability or quantum critical (QC) point which separates the paramagnetic Fermi liquid (FL) phase and the magnetic ordered phase with the ordering vector Q at $T = 0$. Substances belonging to this area have been called nearly and weakly ferro- and antiferromagnetic metals. It seems that the magnetism in this area was called first attention by the discovery of weak itinerant ferromagnets such as $ZrZn_2$ and Sc_3In [1]. The Curie–Weiss (CW) magnetic susceptibility observed in these systems posed a serious challenge to theorists, urging them to consider a new mechanism without local moments. For nearly ferromagnetic metals, on the other hand, attention was attracted by a prediction based on the paramagnon theory that the effective mass in the FL regime diverges logarithmically as one approaches the QC point [2, 3].

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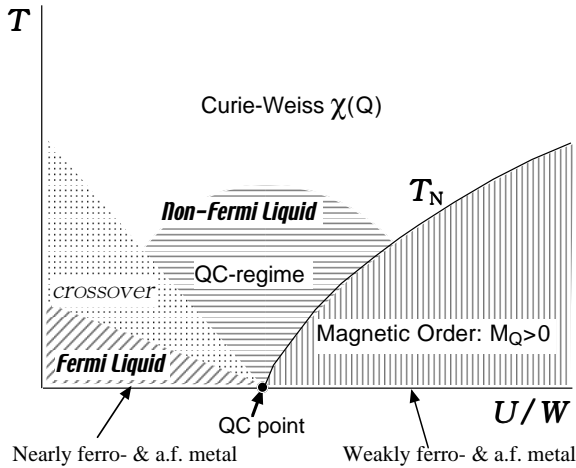


Fig. 1. A schematic phase diagram around the magnetic quantum critical point.

Since then the theory of spin fluctuations has been developed to deal with the problems in the entire area of the phase diagram shown in Fig. 1. The self-consistent quantum mechanical theory of the coupled modes of spin fluctuations or the self-consistent renormalization (SCR) theory not only succeeded in explaining the Curie–Weiss susceptibility of χ_Q but also predicted various singular behaviors of the physical quantities in the QC regime, which have been confirmed by subsequent experimental investigations [4, 5].

The discovery of superconductivity of the materials in this area, heavy electron systems [6], organic conductors [7] and in particular high T_c cuprates [8] has renewed and enormously enhanced the interests in this area.

Upon discovery of the high temperature superconductivity of the cuprates the attention of investigators was first concentrated on the anomalous normal state properties or the so-called non-Fermi liquid properties of these substances where strongly correlated quasi 2-dimensional (2D) electrons in each CuO_2 layer were considered to play a predominant role in conduction [9].

The spin fluctuation theory was extended to cover 2D systems and it was found that the results in the QC and CW regimes were consistent with the observed non-FL behaviors of the cuprates [10]. The experimental results were successfully analyzed in terms of the parametrized SCR theory and the parameter values were estimated.

In the next step the spin fluctuation-induced superconductivity was studied by using the spin fluctuations thus determined quantitatively. Using both weak coupling [10, 11] and then strong coupling theories [12, 13] the superconductivity of $d_{x^2-y^2}$ symmetry was derived with the values of the transition temperature T_c consistent with the experimental results. The d -wave symmetry of the superconducting order parameter of the cuprates was confirmed by experimental investigations in the following years [14].

Subsequent studies on the relation between T_c and the spin fluctuation parameters showed that the value of T_c was roughly proportional to the parameter T_0 indicating the energy spread of the spin fluctuations [15, 16]. This relation may be extended to certain 3D systems and it was shown that the plots of experimental values of T_c against T_0 for cuprates and heavy electron systems came around a straight line [17]. This result suggests the common origin of superconductivity in these systems.

Theoretical studies using the Hubbard and the d - p models have been advanced not only for the cuprates [18–25] but also for the 2D organic superconductors [26–28] by using the fluctuation exchange (FLEX) approximation which may be regarded as the simplest self-consistent theory for the coupled modes of spin fluctuations. The results turned out to be successful indicating that the Hubbard model with proper transfer matrices and a proper electron occupation is a good model for these systems.

A phase diagram is then calculated within the same approximation in a parameter space of the Hubbard model containing the parameter values for both the cuprates and the organic superconductors. It was found that both of them belong to a continuous region of a superconducting phase, indicating their common physical origin [29].

The theoretical approaches discussed so far are the one around the magnetic instability or QC point, which belongs to the intermediate coupling regime. As will be discussed later in this article there have been growing pieces of evidence to indicate that the high T_c cuprates are in the intermediate coupling regime and the spin fluctuation theory is reasonably applied to the main part of the problem.

At this point we would like to mention the pseudo-gap phenomena observed in the under-hole-doped cuprates as a still remaining outstanding subject in the high T_c problems. Although the physics of doped Mott insulators in the entire concentration range, including the pseudo-gap phenomena, seems still to be worked out theoretically, it is fortunate that at least a substantial part of the SC phase of the cuprates is around the QC regime and is described essentially in terms of the theories of spin fluctuations.

In the following sections we summarize very briefly the developments in the theory of spin fluctuations as applied to magnetism and superconductivity and discuss to what extent we now understand these problems.

2. Development of the spin fluctuation theory

Quantum critical and non-Fermi liquid behaviors

The dynamical susceptibility around the magnetic instability for the state with the ordering vector Q may be expressed for small q and ω/q^θ as follows:

$$\frac{1}{\chi(Q+q, \omega)} = \frac{1}{\chi(Q)} + Aq^2 - iC \frac{\omega}{q^\theta}, \quad (1)$$

where $\theta=1$ and 0 for $Q=0$ and $Q \neq 0$, respectively. For non-interacting systems A , C and $1/\chi_0(Q)$ are calculated from the given band structure [30] and RPA result is simply given by $1/\chi_{\text{RPA}}(Q) = 1/\chi_0(Q) - 2U$, U being the on-site interaction constant and the susceptibility is defined without a factor $4\mu_B^2$.

In the self-consistent renormalization theory the renormalized values of A and C usually stay constant around the magnetic instability while the temperature dependence of $1/\chi(Q)$ is strikingly renormalized by the mode-mode coupling effects, *i.e.*, from the Sommerfeld expansion to the quantum critical and Curie-Weiss behaviors.

Leaving derivations of the theory for Ref. [4,5] we show here only the equations for $\chi(Q)$.

$$\begin{aligned} \frac{1}{\chi(Q)} &= \frac{1}{\chi_0(Q)} - 2U + \frac{5}{3}F_Q \sum_{\alpha=x,y,z} m_\alpha^2, \\ m_\alpha^2 &= \frac{2}{\pi} \int_0^\infty d\omega \left(\frac{1}{2} + \frac{1}{e^{\omega/T} - 1} \right) \sum_q' \text{Im} \chi_\alpha(Q+q, \omega), \end{aligned} \quad (2)$$

where F_Q is the mode-mode coupling constant for those with the wave vectors around Q . Eqs. (1) and (2) should be solved self-consistently for $1/\chi(Q)$.

For convenience we introduce here a reduced inverse susceptibility and the following parameters:

$$\begin{aligned} y &= \frac{1}{2T_A \chi(Q)}, \quad (3) \\ T_A &= \frac{A q_B^2}{2}, \quad T_0 = \frac{A q_B^{2+\theta}}{C 2\pi}, \\ y_0 &= \frac{1}{2T_A \chi(Q, T=0)}, \quad \text{for a paramagnetic ground state} \\ &= -\frac{F_Q p_Q^2}{8T_A}, \quad \text{for a magnetically ordered ground state} \\ y_1 &= \frac{5F_Q T_0}{T_A^2}, \quad (4) \end{aligned}$$

where p_Q is the ordered moment in μ_B per magnetic atom, $q_B = \left(\frac{2D\pi^{D-1}}{v_0} \right)^{\frac{1}{D}}$ is the effective Brillouin zone boundary vector, D dimensionality and v_0 is the volume per magnetic atom. The dynamical susceptibility is now written as:

$$\frac{1}{2T_A \chi(Q+q, \omega)} = y + x^2 - i \frac{\nu}{x^\theta},$$

with

$$\nu = \frac{\omega}{2\pi T_0}, \quad x = \frac{q}{q_B}, \quad (5)$$

and

$$y = y_0 + \frac{D}{2} y_1 \int_0^{x_c} dx x^{D+\theta-1} \left[\ln u - \frac{1}{2u} - \psi(u) \right],$$

with

$$u = \frac{x^\theta (y + x^2)}{t}, \quad t = \frac{T}{T_0}. \quad (6)$$

We summarize the results of this theory as follows [4, 5]:

- (1) $\chi(Q)$ obeys the Curie–Weiss law with the origin different from the traditional local moment mechanism.
- (2) The quantum critical behaviors are obtained as shown in Table I. The 3D and 2D results were obtained in 1970's and 1990's, respectively. Many of the results were confirmed by experimental investigations from 1970's to 1990's. Recent renormalization group studies of the same problem lead to the same QC indices as the above results [31, 32].
- (3) The Curie and Néel temperatures of 3D magnets are given by

$$\begin{aligned} T_C &= 0.1052 p^{3/2} T_A^{3/4} T_0^{1/4}, \\ T_N &= 0.1376 p_Q^{4/3} T_A^{2/3} T_0^{1/3}. \end{aligned} \quad (7)$$

These results were confirmed qualitatively (chemical and physical pressure dependence) and quantitatively by experimental investigations since 1970's.

- (4) Expressions for various physical quantities with the 4 SCR parameters are given and are applicable to a wide range of temperature including both QC and CW regimes.

For early investigations on transition metals and their compounds we refer to [4]. Recently non-Fermi liquid properties of certain heavy f -electron systems are investigated intensively and are analyzed in terms of the QC spin fluctuation of 3D or 2D character depending on substances [33].

TABLE I

Quantum critical behaviors of physical quantities.

	ferromagnet ($Q = 0$)		antiferromagnet	
	3D	2D	3D	2D
χ_Q^{-1}	$T^{4/3} \rightarrow CW$	$-T \ln T \rightarrow CW$	$T^{3/2} \rightarrow CW$	$\frac{-T}{\ln T} \rightarrow CW$
$\frac{C_m}{T}$	$-\ln T$	$T^{-1/3}$	const. $-T^{1/2}$	$-\ln T$
$\frac{1}{T_1}$	$T \chi$	$T \chi^{3/2}$	$T \chi_Q^{1/2}$	$T \chi_Q$
R	$T^{5/3}$	$T^{4/3}$	$T^{3/2}$	T

3. Anomalous normal state properties of the high T_c cuprates

From the beginning of investigations the anomalous or non-Fermi liquid properties in the normal state of the cuprates were represented by the T -linear electrical resistivity in a wide temperature range, anomalous temperature dependences of the Hall coefficient R_H and NMR T_1 , R_H and $1/T_1 T$ showing the Curie–Weiss behaviors, and the ω -linear relaxation rate of the optical conductivity, *etc.* In an early stage of investigations all of these properties except for the Hall coefficient were successfully analyzed in a consistent manner in terms of the above discussed spin fluctuation theory. The phenomenological parameters of the theory were determined from analyzes of the resistivity and T_1 . By using thus determined dynamical susceptibility a parameter free comparison between theory and experiment were performed successfully on the optical conductivity of $\text{YBa}_2\text{Cu}_3\text{O}_7$ [34].

The Hall effect was studied recently by using the Kubo–Eliashberg formalism on the Hubbard model with appropriate transfer matrices and electron occupations [35]. The RRPA (renormalized random phase approximation) or FLEX approximation was used for the spin fluctuations and the importance of the vertex corrections in dealing with strongly anisotropic scatterings was emphasized. The results for R_H were consistent with experimental results both for the hole- and electron-doped cuprates; R_H in the former is positive and shows a Curie–Weiss behavior while the latter decreases with lowering temperature from a positive value at high temperature and changes sign, the decrement showing a Curie–Weiss behavior.

In the second stage of investigations the pseudo-gap phenomena observed in under-hole-doped cuprates have called attention of investigators. This may be regarded as low temperature corrections to the non-FL behaviors

as discussed in the first stage. We will discuss this problem later in this article and now move to the problem of superconductivity induced by the spin fluctuations.

4. Superconductivity induced by antiferromagnetic spin fluctuations

4.1. Theories using the parametrized spin fluctuations

Since the anomalous normal state properties were explained by the spin fluctuation theory and the dynamical susceptibilities were estimated quantitatively from analyzes of the experimental results, next problem is naturally to see if the same spin fluctuations can explain the high T_c superconductivity of the cuprates.

It was shown in earlier studies by using a weak coupling theory [36,37] and a strong coupling theory [18] that the antiferromagnetic spin fluctuations can give rise to a d -wave superconductivity. The spin fluctuation induced superconductivity of the cuprates were studied first by using the weak coupling theory [10,11] and then the strong coupling theory [12,13] which is more appropriate. The results showed the $d_{x^2-y^2}$ symmetry of the order parameter and values of T_c consistent with the observed values.

After the unconventional d -wave symmetry of the order parameter was confirmed experimentally the relations between the spin fluctuation parameters and T_c were investigated. The most remarkable result is that T_c is roughly proportional to T_0 , the energy spread of the spin fluctuations. Dependences of T_c on the other parameters, T_A , y_0 , and doping concentration are relatively weak.

The effect of dimensionality was also studied. In favorable cases the values of T_c can be comparable in 3D and 2D systems and are roughly proportional to T_0 , although the dependences of T_c on the other parameters are much more significant in 3D than in 2D systems and 2D seems to be generally more favorable than 3D for superconductivity [17,38]. In the same context FLEX approximation was applied to a nearly half-filled Hubbard model with a simple cubic lattice and the calculated T_c was found to be extremely small [39]. A more recent study of a spatially anisotropic Hubbard model with a transfer parameter interpolating between 2D and 3D systems show that T_c varies only slowly on the 2D side of the parameter values and then decreases rapidly as one approaches the cubic limit [40].

We show in Fig. 2 the plots of experimental values of T_c against T_0 for cuprates and heavy electron superconductors [17]. The plots come around a straight line, suggesting the common origin of superconductivity in these groups of systems.

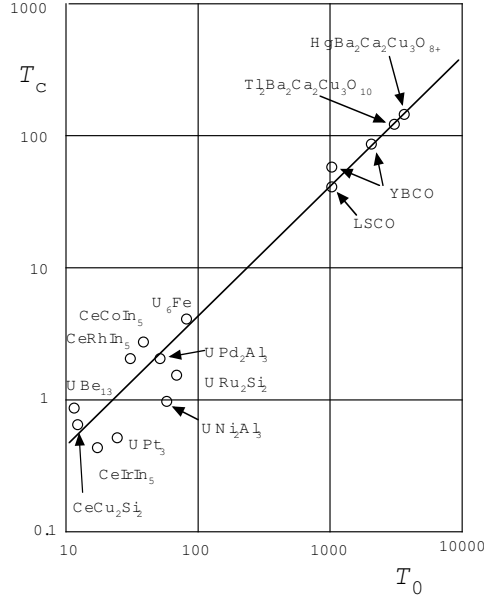


Fig. 2. Transition temperature of unconventional superconductors plotted against T_0 , the characteristic temperature indicating the energy spread of spin fluctuation.

4.2. Theories based on the microscopic models

Fully microscopic calculations of spin fluctuation-induced superconductivity were carried out on the Hubbard and the $d-p$ models by using the FLEX approximation [18–22,24,25], the 3rd order perturbation theory [23], and variational Monte Carlo method [41]. The results may be summarized as follows [5]:

- (1) Superconductivity of $d_{x^2-y^2}$ symmetry is obtained in a fairly large range of intermediate values for U/t . The calculated values of T_c are of reasonable magnitude compared with experiment.
- (2) Similar results are obtained both for the Hubbard and $d-p$ models.
- (3) For the superconducting state, the neutron resonance peak observed in YBCO is explained by the FLEX calculations and by RPA calculations assuming the BCS ground state. Also a peak-dip-hump structure in the one electron spectral density as observed by angle-resolved photoemission experiment is explained.
- (4) Observed differences between the phase diagrams for the electron-doped and hole-doped cuprates are explained at least qualitatively;

a substantially larger concentration range of AF phase and smaller values of T_c in the former as compared with the latter [42, 43]. The calculated ratio of the SC gap and $k_B T_c$ for the former is about half of that in the latter in accord with measured results. This difference may be related with the van Hove singular points which are close to the hot spots, where scatterings due to AF spin fluctuations is particularly strong, in the hole-doped systems but not in the electron-doped systems.

It may be worth while to remark that the FLEX approximation is the simplest possible approach for self-consistently renormalized spin fluctuations and the above success strongly favors the spin fluctuation mechanism for the superconductivity in the cuprates. It is also remarkable that the Hubbard model in the intermediate coupling regime seems to be a good model for the cuprates if one chooses the transfer matrices so as to reproduce the observed Fermi surface for each substance.

5. 2D-organic superconductors and the high T_c cuprates

An extended phase diagram for the Hubbard model

An organic system κ -(BEDT-TTF)₂CuN(CN)₂Cl is an antiferromagnetic insulator at ambient pressure. Under applied pressure it undergoes a weak first order transition, at $p = 200$ bar, into a metallic state which shows superconductivity below $T_c = 13$ K. To a good approximation this substance is considered to be described by a half-filled Hubbard model consisting of anti-bonding dimer orbitals arranged in a square lattice. As for the transfer integrals we may take $-t$ between the nearest neighbors and t' between the second neighbors in one of the diagonal directions.

This model was studied with the FLEX approximation and the results showed the superconducting order parameter of $d_{x^2-y^2}$ symmetry and reasonable values for T_c [26–28, 44]. Since the approach here is just the same as the one discussed in the preceding section for the cuprates, the common success naturally indicates the same origin of superconductivity in these systems.

The above results are of particular interest since the organic compound is found in the metallic side of the Mott transition while the high T_c cuprates are doped Mott insulators. The former is clearly in the intermediate coupling regime and existing approaches from the strong coupling limit does not seem to work.

In order to see the situations more clearly a phase diagram was calculated in a parameter space of the Hubbard model taking the following quantities for the 3 axes of the parameter space: U/t , t'/t and $n - 1$, n being the number of electrons per site [29]. The FLEX approximation was employed

and the results were extrapolated to $T = 0$ with the use of Padé approximants. The calculated phase diagram is shown in Fig. 3. We see that the superconducting states of cuprates and those of the organic compounds are found in the same connected region of a superconducting phase in this phase diagram.

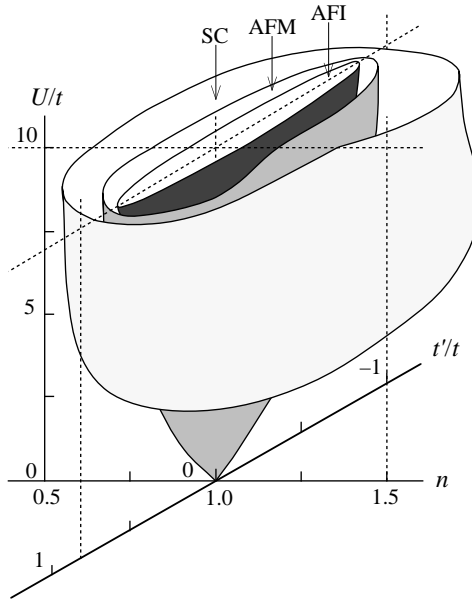


Fig. 3. The phase diagram of a nearly half-filled Hubbard model in a three dimensional parameter space: U/t - t'/t - n . Superconducting (SC) and antiferromagnetic (AFM) instability surfaces are indicated. The boundary between antiferromagnetic metal (AFM) and insulator (AFI) should depend on the impurity potential and is sketched here just to show general idea of its location.

6. Magnetic instability vs. Mott transition

Which is more important for high T_c ?

Among the mechanisms of superconductivity in strongly correlated electron systems the one for the heavy electron systems is considered mainly due to the spin fluctuations, although additional contributions of orbital and charge fluctuations are under investigation [45]. For the 2D organic compounds the only mechanism explicitly worked out so far seems to be the spin fluctuation mechanism.

For the high T_c cuprates, on the other hand, many different mechanisms have been proposed. In addition to the approaches as discussed in the above, various approaches from the strong coupling limit based on the t - J model

have been pursued extensively [46, 47] Since the t - J model is an approximation to the Hubbard model from the strong coupling limit it may be pertinent here to discuss on the phase diagrams of the Hubbard model and see where in the parameter space the high T_c cuprates are really located.

Let us first look at a phase diagram of a half-filled Hubbard model including the magnetic QC point, an antiferromagnetic phase and a metal to insulator transition, Fig. 4. Appropriate ways of approaches for different sections of the phase diagram are also indicated in the figure.

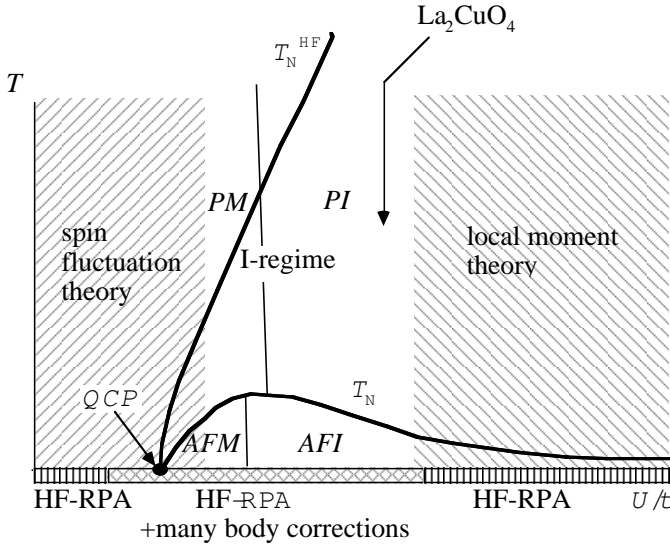


Fig. 4. A sketch of the phase diagram for the half-filled Hubbard model. Appropriate theoretical approaches are indicated in the corresponding sections of the phase diagram.

We first emphasize that in the ground state the Hartree–Fock (HF)–RPA is correct in the strong coupling regime where the ordered moment is enough polarized so that the correlation effect is insignificant; two electrons with opposite spins seldom encounter with each other. The spin wave dispersion calculated by RPA in the antiferromagnetic ground state agrees, in the limit of small t/U , precisely with the one calculated from the Heisenberg model with the Anderson kinetic super-exchange interactions [4]. Since the HF–RPA is known to be correct in the weak coupling limit it should make a fair interpolation between the strong and weak coupling limits in the ground state. In the intermediate coupling regime, where the electron–electron correlations are most significant, it is a reasonable approach to start from HF–RPA and to make many body corrections. The self-consistent theory of renormalized spin fluctuations may be an example of possible approaches.

At finite temperatures, however, HF-RPA is not at all a good approximation in any regimes of present interest. For the Mott insulator phase with large U/t the local moment model with the Anderson kinetic super-exchange (t/U expansion) is valid while for weak and intermediate U/t just covering the antiferromagnetic instability the SCR spin fluctuation theory is considered to be an adequate approach. For the intermediate regime between these two including the Mott transition no really satisfactory approach at finite temperatures is known so far.

We note that 2D organic superconductors are on the metallic side of the first order Mott transition and is considered to be located around the hidden antiferromagnetic instability and thus are expected to be well treated by the spin fluctuation theory.

Next we discuss doped Mott insulators. If we neglect the impurity potential the ground state of a doped Mott (antiferromagnetic) insulator is an antiferromagnetic metal. On doping we first have small Fermi surfaces which expand with increasing doping concentration at the expense of reduced gap area. Then we have a large Fermi surface with small gap area and finally have a paramagnetic metal through the QC point where the AF gap just vanishes.

As is seen in Fig. 5 the magnetic instability or QC point should make a continuous line in a U/t against n plane extending from the half-filled ($n = 1$) case with relatively small U/t toward larger U/t and $n \neq 1$. The spin fluctuation theory is an approach along the QC line from the side of weaker coupling and is expected to cover at least the intermediate coupling regime. As a matter of fact the values for U/t used in the above calculations are around $4 \sim 8$ and thus U is about $0.5 \sim 1$ times the band width. This is consistent with the value estimated from photoemission experiments [48].

According to recent analyzes of the spin wave energy dispersion in antiferromagnetic La_2CuO_4 , a parent of high T_c cuprates, as measured by neutron scattering studies [49] the dispersion in the entire Brillouin zone is well reproduced by an RPA calculation on the Hubbard model with $U/t = 6$ [50]. In order to get a reasonable fit with the local moment model one needs to consider the higher order terms in the t/U expansion including the ring exchange terms [49]. This fact seems to evidence the intermediate coupling nature of the cuprates and supply another support for the applicability of the spin fluctuation theory to the high T_c problems and explain the reason for its success.

On the other hand the approaches based on the t - J model with a few terms of Anderson super-exchange interactions might not even cover the parent insulator state. Applicability of this approach as an approximation to the Hubbard model may also be limited to a very low doping concentration range. For larger doping concentrations it may take us into a different world from the one described by the Hubbard model.

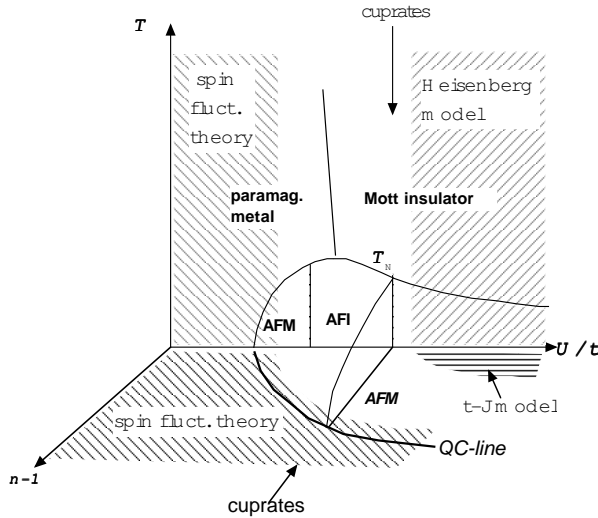


Fig. 5. A sketch of the phase diagram for the nearly half-filled Hubbard model. Appropriate approaches are indicated in the corresponding sections.

7. Pseudo-gap phenomena

Pseudo-gap phenomena were first observed in NMR T_1 measurements on underdoped YBCO [51, 52] as a pseudo-gap in spin excitations. The measurements were then extended to transport properties, one electron spectral density as observed by ARPES and tunneling experiments and various theoretical mechanisms were proposed [53].

According to the FLEX calculations T_c generally tends to increase with lowering doping concentration until it reaches the AF phase boundary calculated introducing weak 3-dimensionality. This behavior is consistent with the experimental results on the electron-doped cuprates. For the hole-underdoped cuprates, however, experimental results seem to suggest that there is some additional mechanism to suppress both the AF and SC orderings, giving rise to the pseudo-gap at the same time.

Among various possibilities proposed so far one natural direction from the present point of view may be to consider superconducting fluctuations (p - p channel) in addition to the AF spin fluctuations (p - h channel). The SC fluctuation is expected to be particularly significant in 2D systems with short coherence lengths. Here we refer to an investigation reported recently [54-56]. The SC fluctuations are taken into account within the t -matrix approximation together with the FLEX spin fluctuations and the equations are solved self-consistently with still further approximation of expanding the t -matrix in q^2 and ω near the SC critical point. The results include many attractive features:

- (1) In the under-hole-doped regime T_c decreases from the FLEX value and the value of T_c decreases with lowering doping concentration. The AF phase tends to be suppressed at the same time.
- (2) Pseudo-gap behaviors in the one electron spectral density and reduction of the NMR relaxation rate $1/T_1T$ are obtained.
- (3) Observed pseudo-gap behaviors in the transport properties: electrical resistivity, Hall coefficient, magneto-resistance, thermo-electric power, and the Nernst coefficient are all explained consistently.
- (4) Calculated effects are significant in the hole-underdoped cuprates but not in the electron-doped cuprates in accord with experiment.

In order to make this scenario convincing it is desired to extend the theory to still lower doping concentrations and clarify the phase diagram near the AF insulator phase.

8. Conclusion

We have summarized the present status of the theories of the spin fluctuation mechanism in explaining anomalous or non-Fermi liquid behaviors and unconventional superconductivity in strongly correlated electron systems around the magnetic quantum critical point, referring to the high T_c cuprates, 2D organic compounds and heavy electron systems. So far the spin fluctuation theories seem to be successful in explaining at least essential parts of the problems, indicating in particular that the spin fluctuation is the common origin of superconductivity in these systems.

As for the high T_c cuprates at least the central part of the SC phase seems to be approached by the spin fluctuation theory. It was fortunate that the high T_c cuprates are in the intermediate coupling regime and are close to the magnetic instabilities. It still remains to describe all the hole-underdoped regime lying between the AF insulator phase and the optimal concentration regime, where the pseudo-gap phenomena are still controversial indicating the need for another mechanism in addition to the antiferromagnetic spin fluctuations.

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