Review

Developments of the theory of spin fluctuations and spin fluctuation-induced superconductivity

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Abstract: Theory of spin fluctuations as developed in the past 30 years have played important roles in the theory of magnetism in metals, particularly in elucidating the properties around the magnetic instability or quantum critical points. Recently the theory has been extended to deal with the spin fluctuation-mediated superconductivity with anisotropic order parameters in strongly correlated electron systems. These theoretical developments are briefly reviewed and the high temperature superconductivity of cuprates and organic and heavy electron superconductors are discussed in the light of these theories.

Key words: Spin fluctuation; superconductivity; high-$T_c$ cuprates; itinerant electron magnetism; magnetic quantum critical point.

1. Introduction. Studies of magnetism have a long history of more than 2000 years. However, its real understandings came after the advent of quantum mechanics in the 20th century. Even with quantum mechanics understanding of this problem did not proceed quite straightforwardly. This is because the electrons which play predominant roles in strong magnetism such as in ferro- and antiferromagnetic metals are strongly correlated and the traditionally successful mean field theory is quite insufficient. The well-known controversy between localized and itinerant electron pictures lasted for a long time and this and many other problems have been settled from time to time, but the problems have not entirely been solved yet.

In contrast superconductivity was discovered early in the 20th century and the problem was solved in the middle of the century by the Bardeen-Cooper-Schrieffer (BCS) theory where the phonon-mediated attractive interaction between electrons gave rise to a condensation of paired electrons and the use of a mean field approximation was justified. Here magnetism was considered to be an obstructive factor of superconductivity.

Recent discoveries of superconductivity in heavy electron systems, organic systems and in high-$T_c$ cuprates have opened a new area of research where superconductivity is considered to have a magnetic origin. Since then the problem of magnetism and superconductivity in strongly correlated electron systems has formed one of the most active area of research in physics.

In this article we first discuss very briefly some developments in the theory of magnetism of metals leading to the spin fluctuation theory and then go on to discuss recent theories of spin fluctuation-mediated superconductivity which are now considered to be appropriate as applied to high-$T_c$ cuprates, organic and heavy electron superconductors.\(^{1)}\)

2. Historical developments of the theory of itinerant electron magnetism leading to the theory of spin fluctuations.\(^{2)}\) There have been two main streams in the developments of the modern theory of magnetism started with the advent of quantum mechanics. One is based on the local moment model of Heisenberg 1928, a quantum mechanical innovation of the classical Langevin-Weiss theory which assumes an atomic magnetic moment of a fixed size. The other, started with Bloch 1929, is based on the itinerant electron
model or the band theory of electrons in metals. These two models have mutually opposite starting points. The starting picture of the former consists of electrons localized on each atom of a crystal while that of the latter consists of itinerant electrons specified by their wave vectors.

The Heisenberg model was successful in describing representative physical properties of ferromagnets; the second order phase transition between ferro- and paramagnetic states at the Curie temperature $T_C$ and the Curie-Weiss magnetic susceptibility: $\chi = C / (T - T_C)$ above $T_C$, etc. This model was then extended to include the exchange interactions of wider spatial range and of negative sign. Various types of magnetic anisotropy were also taken into account. We now know that wide variety of magnetic ordered structures, including antiferromagnetism, ferrimagnetism, helimagnetism, spin-canted weak ferromagnetism, etc., their temperature dependencies and phase transitions are described by using extended Heisenberg spin Hamiltonians.

The Heisenberg model was first derived by extending the Heitler-London molecular binding theory to solids neglecting the overlap integrals and this neglect and the theoretical foundation of the model itself was questioned later. In 1959 Anderson developed a theory of magnetic insulator compounds, giving a firm ground to the local moment model with the superexchange interaction of mostly antiferromagnetic sign. This theory starts with recognition that all the magnetic insulator compounds are the Mott insulators, where the electrons are localized owing to strong electron-electron correlations. The simplest model for this problem consists of the transfer integral $t$ between neighboring atomic orbitals and the intra-atomic interaction $U$. The number of electrons is one per atom. For infinite $U$, every electron is localized on an atom or a lattice site and the system is an insulator. For small $t/U \ll 1$ the system continues to be an insulator and an expansion in $t/U$ is theoretically justified. Anderson derived a now famous kinetic superexchange interactions from the second order terms. Additional consideration of the spin-orbit coupling in the degenerate orbital model leads to all kinds of magnetic anisotropy in the spin Hamiltonian.

After productive investigations on the local moment model for nearly a century, a rich variety of magnetic structures, their temperature dependencies and the phase transitions between various phases were elucidated. Investigations along this stream still continue with special interests in low dimensional systems and frustrated spin systems where the ground state is hardly ordered magnetically.

The itinerant electron model of ferromagnetism was developed by Bloch, Salter and Stoner on the basis of the mean field theory. Bloch studied possible ferromagnetism of an electron gas by using the Hartree-Fock (HF) mean field theory. His conclusion was that an electron gas can be ferromagnetic when its density is lower than a certain critical value. Wigner pointed out the importance of electron-electron correlations, which were neglected in the HF theory, and concluded that the electron gas cannot be ferromagnetic. Since then the tight-binding model with onsite interactions has been used widely to discuss itinerant electron magnetism. Salter discussed ferromagnetism of Ni by using a calculated band structure and Stoner developed a theory of ferromagnetism at finite temperatures, both on the basis of the HF mean field theory. The Stoner theory as elaborated by Wohlfarth and others was applied to a number of ferromagnetic metals. We now realize that the Stoner theory was not successful after all in describing finite temperature properties of ferromagnetic metals in any consistent way. The most famous two drawbacks were (1) too high calculated values of $T_C$ as compared with experiment and (2) impossibility of describing the Curie-Weiss susceptibility under realistic conditions.

In an early stage of investigations both the localized and the itinerant electron models were applied to discuss ferromagnetic metals, since a picture of localized d-electrons was supported by many investigators. Theoretical consequences of these models have merits and demerits in an opposite way. For example the Curie-Weiss susceptibility is a well-known consequence of the former while the latter could not explain it within the mean field theory as mentioned already. On the other hand, fairly large low temperature $T$-linear specific heat observed in ferromagnetic metals is naturally explained in the latter but not in the former. Thus serious controversy between localized and itinerant pictures continued since an early stage for at least 30 years.

Around 1960’s a widely accepted point of view, after the long controversy, was that the magnetic insulator compounds and rare earth magnets are described in terms of the localized electron model while the ferromagnetic d-electron metals should be described on the itinerant electron model with the approximation method beyond the mean field level, properly taking account of the effects of electron-electron correlations. One of the clearest motivations for this consensus was the successful experimental observations of the d-electron Fermi surfaces in ferromagnetic Fe and Ni and their
good comparisons with the results of band theoretical calculations.

Thus it became necessary to improve over the HF theory of itinerant electron magnetism by taking account of electron-electron correlations. A natural way in this direction may be to take into account spin density fluctuations neglected in the HF theory. The random phase approximation (RPA) theory for the dynamical susceptibility was developed successfully to describe spin waves in the ferromagnetic ground state but it could not improve the above-mentioned drawbacks of the HF theory. This is because the RPA theory deals with the fluctuations around the HF equilibrium state which remains without being renormalized.

A popular theoretical approach in 1960’s was to consider local magnetic moments in metals. This was natural since almost all ferromagnetic metals showed the Curie-Weiss susceptibility above their Curie temperatures. Nevertheless they showed very good Curie-Weiss susceptibility in very wide temperature ranges above $T_C$. These experimental results indicated that there is a new mechanism for the CW susceptibility different from the traditional local moment mechanism and gave impact to the study of advancing the theory of spin fluctuations. Before going to discuss this development let us briefly summarize the RPA theory at the preceding stage.

### 3. RPA theory of spin fluctuations

After the mean field theoretical study of itinerant electron ferromagnetism, spin waves and other magnetic excitations were investigated by extending the mean field theory to include dynamical variables. To explain the theory explicitly we use the following simplest tight-binding Hamiltonian with on site electron-electron interactions (Hubbard model), expressed in terms of annihilation and creation operators for the electrons in the local (Wannier) or extended (Bloch) orbitals.

$$
H = \sum_{\sigma \sigma'} \sum_{j l} t_{j l} \sigma \sigma' a_{j l \sigma}^{\dagger} a_{l j \sigma'} + U \sum_j n_{j \uparrow} n_{j \downarrow}
$$

$$
I = U / N, \quad \text{[1]}
$$

where $\sigma$ indicates spin $\uparrow$ or $\downarrow$, $t_{j l}$ is the transfer integral, $\varepsilon_{k\sigma}$ is the energy of an electron in the band, $U$ the intra-atomic or onsite interaction between electrons, and $N$ is the number of atomic sites in the crystal.

The interaction term is approximated by the molecular field common to all the electrons.

$$
I \sum_{\sigma \sigma'} \sum_{q h k} a_{h k \sigma}^{\dagger} a_{q h \sigma'}^{\dagger} a_{k h \sigma} a_{q h \sigma'} \rightarrow I \sum_{\sigma \sigma} \sum_{h k} a_{h k \sigma}^{\dagger} \left( a_{h k \sigma} a_{h k \sigma'} - \frac{1}{2} \right) - I N_{\uparrow} N_{\downarrow}
$$

$$
= I \sum_{\sigma \sigma} \sum_{h k} a_{h k \sigma}^{\dagger} a_{h k \sigma} N_{\sigma} - I N_{\uparrow} N_{\downarrow}. \quad \text{[2]}
$$

We now define the magnetization $M$ and the total number of electrons $N$ as follows:

$$
2M = N_{\uparrow} - N_{\downarrow}, \quad N = N_{\uparrow} + N_{\downarrow}. \quad \text{[3]}
$$

The HF Hamiltonian is given by

$$
H_{\text{HF}} = \sum_{\sigma \sigma} \sum_{h} (\varepsilon_{h} + \Delta \alpha_{h} n_{h \sigma}) a_{h \sigma}^{\dagger} a_{h \sigma}, \quad \Delta = IM, \quad \text{[4]}
$$

except for a constant term and $2\Delta$ is the exchange splitting of the up and down spin bands. We sketch in Fig. 1 the densities of states for a split band ferromagnet at $T = $
The Fermi surfaces for the up and down spin bands are sketched in Fig. 2 for an electron gas like band, where an electronic spin flip excitation is indicated; an electron with the wave vector $k + q$ and spin down is excited to the state with the wave vector $k$ and spin up. Thermal spin flip excitations will reduce the magnetization $M$ until it vanishes at $T_C$. In the mean field theory the energy of a spin flip excitation (Stoner excitation) is given by

$$\omega_{\text{Stoner}}(k, q) = 2\Delta + \varepsilon_k - \varepsilon_{k+q}. \quad [5]$$

In order to study the spin flip excitation spectrum more generally we consider an equation of motion for the following spin flip excitation operator:

$$S_i(k, q) = a_{k+q}^\dagger a_k^\dagger,$$  \quad [6]

with the additional oscillating external field Hamiltonian:

$$H_{\text{ext}} = \sum_k S_i(k', -q) h_i(q),$$

where $h_i(q)$ is $2\mu_B$ times the oscillating external field. After calculation we get

$$\frac{\partial}{\partial t} S_i(k, q) = [S_i(k, q), H + H_{\text{ext}}]$$

$$= (\varepsilon_{k+q} - \varepsilon_k - 2\Delta) S_i(k, q) + \sum_k (n_{k+q} - n_k) S_i(k, q)$$

$$+ i \sum_k \sum_{q'} \left(1 - \delta_{k+q, k+q'} \right) \langle a_{k+q'}^\dagger a_k^\dagger \rangle S_i(k, q)$$

$$- i \sum_{q'} \sum_k \left(1 - \delta_{k+q', k+q} \right) \langle a_{k+q}^\dagger a_k^\dagger \rangle S_i(k, q). \quad [7]$$

Here and in what follows we use the energy units for $\omega$ and $T$, setting $k_B = 1$ and $m = 1$. The dynamical susceptibility as defined by $\chi_{\Delta}(q, \omega) = -\sum_k \langle S_i(k, q) \rangle / \hbar(q)$ is calculated from this equation assuming $h_i(q)$ to oscillate as $\exp(i\omega t)$. Taking only the first line of this equation gives

$$\chi_{\Delta}(q, \omega) = \sum_k \frac{\langle n_{k+q} \rangle}{\varepsilon_k - \varepsilon_{k+q} + 2\Delta - \omega}. \quad [8]$$

The imaginary part of this expression gives the intensity spectra of the Stoner excitations where the electrons and holes as shown in Fig. 2 move independently in the uniform molecular field.

The second line in eq. [7] gives the effect of scatterings of the electron-hole pairs with the restriction of fixed momentum transfer or total momentum conservation. The last two lines are quadratic in the density operators and gives the effects of coupling among the different modes (wave vector components) of density fluctuations. Neglecting these lines leads to the random phase approximation (RPA). We discuss here the consequences of RPA and the effects of the third line
Taking a statistical average of eq. \[7\] neglecting the last two lines and summing over \(k\) gives

\[
\sum_k \langle S_k, q \rangle = -\chi_{k0}^{+}(q, \omega) b(q) + i \chi_{k0}^{+}(q, \omega) \sum_k \langle S_k, q \rangle,
\]

and the RPA dynamical susceptibility is given by

\[
\chi_{RPA}^{+}(q, \omega) = \frac{\chi_{k0}^{+}(q, \omega)}{1 - i \chi_{k0}^{+}(q, \omega)}.
\]

We see that the RPA susceptibility is generally enhanced from the mean field value eq. \[8\]. The spin flip excitation intensity spectra are given by the imaginary part of the dynamical susceptibility. We show in Fig. 3a and b the calculated intensity contours for the RPA and Stoner excitations, respectively. The excitation spectra are dramatically different between two approximations. The low energy part of the excitations is strongly enhanced in RPA compared with Stoner. Fig. 3a shows the spin wave excitations in the area where the Stoner excitations do not exist. The spin wave is the bound state of an electron-hole pair.

We now see why the Stoner mean field theory, which makes use of the spin excitation spectra Fig. 3b, gives too high values for \(T_c\). One had better use the RPA(Fig. 3a) rather than the Stoner(Fig. 3b) excitations in calculating finite temperature properties of itinerant ferromagnets.

Improvements of the theory along this direction was initiated in mid-1960’s for nearly ferromagnetic metals \(^4\)-\(^6\) and theories for ferro- and antiferromagnetic metals around the magnetic instability or quantum critical point (QCP) were developed in 1970’s \(^7\)-\(^9\).

4. Theory of spin fluctuations around the magnetic instabilities (QCP). We start this section with the following equation for the free energy (as a function of magnetization) expressed in terms of spin fluctuations or dynamical susceptibilities:

\[
F(M, T) = F_{HF}(M, T) - \frac{1}{2\pi} \int \frac{d\omega}{2\pi} \int dI \text{Im} \left[ \chi_{Mi}^{+}(q, \omega + i\delta) - \chi_{Mi}^{0}(q, \omega + i\delta) \right].
\]

where \(F_{HF}\) is the Hartree-Fock free energy and \(\chi_{Mi}^{0}(q, \omega + i\delta)\) is the transverse dynamical susceptibility of a ferromagnet with magnetization \(M\) and interaction.
constant $I, s$ being $\rightarrow + 0$. This expression is derived for the Hubbard model using the coupling constant integral and is an exact expression. In practice we can calculate the free energy approximately by using various approximations for $\chi_{MF}(q, \omega + i\epsilon)$.

When the mean field susceptibility eq. [8] is used we get $F_{HF}$. In mid-1960's the low temperature specific heat of nearly ferromagnetic (paramagnetic) metals were studies by using RPA susceptibility eq. [9] in eq. [10] for $M = 0$. In this problem components of spin fluctuations with small $q$ and small $\omega/q$ are important and the following expansion form is useful:

$$\frac{1}{\chi_{RPA}(q, \omega)} = \frac{1}{\chi_{HF}(0, 0)} + A_q^2 - iC_q \frac{\omega}{q},$$  \hspace{1cm} [11]

where $A_q$ and $C_q$ can be calculated from the band structure near the Fermi surface. After calculation it was found that the T-linear coefficient $\gamma$ of the specific heat diverges logarithmically as the ferromagnetic instability or quantum critical point (QCP) is approached. Also there was a term proportional to $T^3 \log T$. Magnetic susceptibility $1/\chi = \hat{\delta}^2 F / \delta M^2$ can be calculated by using RPA expressions in eq. [10]. With a long wavelength approximation we get the following expression:

$$\frac{1}{\chi} = \frac{1}{\chi_{HF}} + \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{\mathbf{q}} \{ F_i \text{Im} \chi_{HF}^{\Delta}(q, \omega + i\epsilon)$$

$$+ \{ D_i \omega / q^2 \} \text{Im} \chi_{HF}^{\Delta}(q, \omega + i\epsilon) \} \}.$$  \hspace{1cm} [12]

$F_i$ and $D_i$ are the mode-mode coupling constants (of positive sign) for the long wavelength modes of spin fluctuations. After calculation it was found that the $T^2$ term of $1/\chi$ is strongly enhanced, the coefficient diverging as the QCP is approached.

Subsequently nearly antiferromagnetic metals were studied with the same approximation. We have the following form of expression for the dynamical susceptibility corresponding to eq. [11]. $Q$ is the wave vector specifying the antiferromagnetic spin arrangement.

$$\frac{1}{\chi_{RPA}(Q + q, \omega)} = \frac{1}{\chi_{HF}(Q, 0)} + A_q q^2 - iC_q \omega.$$  \hspace{1cm} [13]

It was shown that neither logarithmic divergence of $\gamma$ nor $T^3 \log T$ term exists near the antiferromagnetic QCP.

When one wishes to extend this approach to weakly ferro- and antiferromagnetic metals through their QCP, we encounter serious difficulties. For example, let us consider to calculate $T_c$ from eq. [12] as a point of vanishing $1/\chi$ in the paramagnetic phase. Since the second term of the right hand side of eq. [12] is positive, $T_c$ must be lower than $T_{HF}$. On the other hand the expression for $\chi_{HF}(q, \omega)$ shows paramagnetic behaviors only above $T_{HF}$ and thus eq. [12] is meaningful only above $T_{HF}$; one cannot reach $T_c$ which is lower than $T_{HF}$. In order to calculate $T_c$ it is essential to use an expression for the dynamical susceptibility whose long wavelength and static limit is consistent with the $1/\chi$ on the left hand side, i.e. the result of calculation. This self-consistency requirement is violated when one uses RPA expression in eq. [10].

One simple way to remedy this discrepancy is to use in eq. [12] the following form of the dynamical susceptibility:

$$\frac{1}{\chi(q, \omega)} = \frac{1}{\chi} + A_q q^2 - iC_q \omega / q.$$  \hspace{1cm} [14]

in place of $\chi_{HF}(q, \omega)$ in eq. [11]. This way of thinking underlies the first derivation of the self-consistent renormalization (SCR) theory of spin fluctuations where the free energy and the dynamical susceptibility are renormalized self-consistently. We mention here two remarkable results of the SCR theory. One is a substantial (order of magnitude) reduction of $T_c$ from its HF value. Another is the Curie-Weiss susceptibility in a very wide temperature range above $T_c$, and the mechanism is different from the traditional one with local moments. We show an early result of calculation in Fig. 4a, together with typical experimental results in Fig. 4b.

Various alternative derivations of the SCR theory were presented in subsequent years, including the classical and quantum mechanical functional integral methods, a method of equations of motion and corresponding diagrammatical Green's function methods and a phenomenological Ginzburg-Landau type approach, etc. The equations of motion and diagrammatical theories, as a matter of fact, do not make use of the long wavelength or low frequency approximation and may be regarded as a general theory improved substantially over the HF-RPA theory. With the long wavelength and low frequency approximations the equations reduce to those for the previously derived SCR theory around QCP.

We now summarize the main results of the SCR theory around the QCP. As already mentioned partly, the Curie-Weiss susceptibility for $\chi(Q)$, $Q$ being the spin...
ordering vector, is one of the most important results of the theory. The quantitative value for the Curie temperature is expressed in terms of the parameters $A$, $C$ for the spin fluctuations given by the following dynamical susceptibility:

$$\frac{1}{\chi(Q + q, \omega)} = \frac{1}{\chi_Q} + Aq^2 - iC \frac{\omega}{q^2}. \quad [15]$$

$\theta = 1$ for $Q = 0$ and $\theta = 0$ for $Q \neq 0$, and the value for the saturation moment. These parameters can be measured by neutron inelastic scattering experiments and magnetization measurements. Actual studies were performed for ferromagnetic ZrZn$_2$, MnSi, Ni$_3$Al and antiferromagnetic V$_{2-x}$O$_3$ and the quantitative agreements were quite satisfactory.$^{16-18}$

Another important results were predictions for
anomalous behaviors just at the quantum critical point (QCP). The results are summarized in Table I. Corresponding anomalous behaviors near the QCP are summarized in Table II.

The predictions for three-dimensional systems have been confirmed experimentally since 1970’s. The theoretical results for two-dimensional systems were given in 1990 upon discovery of high-$T_c$ cuprate superconductors, as will be discussed in the following section. These theoretical results were endorsed later with the use of a renormalization group theory.

The phenomenological version of the SCR theory around the QCP has 4 parameters, $A$, $C$, $F_1$ (the mode-mode coupling constant), and the saturation moment or the zero-temperature susceptibility. For convenience we express the 4 parameters of the SCR theory around QCP as follows:

\[
T_A = Aq_B^2/2, \
T_0 = (A/C)q_B^2/2\pi, \
y_0 = 1/2T_A \chi(Q,T = 0), \text{ for a paramagnetic ground state}, \
y_1 = 5F_Q T_0 / T_A^2, \\
\]

where $q_B$ is the effective Brillouin zone boundary vector and $p_Q$ is the ordered moment per atom in the ground state. Various physical quantities were expressed in terms of these parameters and analyses of experimental results were performed with success for a number of weakly and nearly ferro- and antiferromagnetic metals. For details we refer to 1), 2) for interested readers.

5. Discovery of high-temperature superconductors and their anomalous normal state properties—non-Fermi liquid properties. In 1986 J. G. Bednorz and K. A. Muller reported their discovery of superconductivity in doped cuprates $La_{2-x}Sr_xCuO_4$ with a layered perovskite structure below $T_c \sim 40$ K. In the next year C. W. Chu et al. discovered another cuprate superconductor $YBa_2Cu_3O_{6+x}$ with $T_c \sim 90$ K. Since then a number of superconductors have been reported in the same series of layered perovskite structures and the highest value of $T_c$ reached $\sim 150$ K. These substances have been called 'high temperature superconductors' and attracted worldwide attention of an enormous number of investigators. We note that for a long time before this discovery the highest record of $T_c$ was less than 30 K.

The most remarkable characteristics derived from intensive investigations in an early stage were (1) These substances were obtained by doping antiferromagnetic Mott insulators with holes or electrons. We sketch in Fig. 5 a conceptual phase diagram of the doped cuprates. (2) The carriers are confined in each CuO$_2$ layer and have strong two-dimensional character, (3) Physical properties in the normal state (above $T_c$) show anomalous behaviors quite different from those of normal metals or Fermi liquids. These properties have been called non-Fermi liquid properties and were considered to form a key issue to understand the entire problem.

Although a number of theoretical proposals have been presented since an early stage of investigations, we may now focus on two lines of approaches emphasizing the importance of electron-electron correlations and magnetic origin of the superconductivity. One emphasizes the closeness of the systems to the Mott insulator phases while the other put emphasis on their closeness to the antiferromagnetic QCP.

The former line of approaches extend the Anderson theory of the Mott insulators to doped systems, expecting intrinsic non-Fermi liquid behaviors in view of apparent inaccessibility of Mott insulators from the band theory. In practice, so-called $t$-$J$ model is adopted, where carriers move from site to site by transfer integral $t$ strictly avoiding double occupancy of a site. As for the interaction between electrons kinetic super-exchange interaction $-\langle 4t^2/U \rangle \langle \vec{S}_1 \cdot \vec{S}_2 \rangle$ is considered when they come to neighboring sites. This is an approach from the strong coupling limit and is expected after all to be...
connected continuously to the realistic intermediate coupling regime. Enormous efforts have been concentrated on this side of approaches in the past two decades and formalisms and numerical methods have been advanced significantly.\(^{26}\) However, it still seems to be hard with this approach at the present time to make systematic comparisons between theories and experimental results.

On the other hand the other line of approaches, making use of the spin fluctuation theory as discussed in the preceding section, were successful in explaining the non-Fermi liquid properties and the high-\(T_c\) superconductivity consistently. We will thus discuss this approach and its consequences in what follows.

We show some of typical experimental results of the anomalous normal state properties in Fig. 6. (1) Electrical resistivity is linear in temperature \(T\) in a wide range of temperature above \(T_c\). In Fermi liquids (FL) the resistivity is known to have \(T^2\) dependence. (2) The nuclear spin-lattice relaxation rate divided by \(T\) shows temperature dependence of the Curie-Weiss type, while in FL there is primarily no temperature dependence.

Fig. 6. Anomalous physical properties in the normal state of high-\(T_c\) cuprates. (a) electrical resistivity, (b) Hall coefficient, (c) nuclear spin-lattice relaxation rate divided by temperature: \(1/T_1 T\), (d) relaxation rate in the optical conductivity.
dependence. (3) The relaxation rate associated with the optical conductivity has linear frequency dependence, while it is constant in FL. (4) The Hall coefficient shows Curie-Weiss type temperature dependence, while it is constant in FL.

These anomalous behaviors are reminiscent of the quantum critical behaviors as studied for 3-dimensional systems in 1970's. Thus the SCR theory was extended to 2-dimensional antiferromagnetic systems and the results as shown in Table I were just consistent with the above mentioned experimental results (1) and (2).

From analyses of these experimental results the SCR parameters for the spin fluctuations, eq. [16], were estimated and the optical conductivity was calculated quantitatively by using these parameters. The results without adjustable parameter compared well with experimental results.

The Hall coefficients were calculated both for hole- and electron-doped cuprates with the use of the Kubo formula for the conductivity and the FLEX approximation for the spin fluctuation. The results well explained the temperature dependencies of the Hall coefficients, including the opposite trends between hole- and electron-doped systems.

Since the anomalous normal state properties are considered to arise from the effects of antiferromagnetic spin fluctuations it may be natural to expect the same spin fluctuations to mediate superconductivity of high-\( T_c \) cuprates. Investigations along this direction have been reported since around 1990. This subject will be discussed in the following section.

6. Theories of spin fluctuation-induced superconductivity and their application to high-\( T_c \) cuprates and other strongly correlated electron systems. The BCS theory of superconductivity mediated by electron-phonon interactions deals with the simplest case of an isotropic superconducting gap. Although the possibility of an anisotropic gap has been investigated soon after the BCS theory, the first example was discovered as the superfluidity of \( ^3\)He where the triplet pairing with a symmetry of p-wave character is realized. The pairing was considered to be mediated by nearly ferromagnetic spin fluctuations.

In 1979 superconductivity was discovered in a heavy electron system CeCu\(_2\)Si\(_2\), followed by UBe\(_{13}\) and other f-electron systems. The electrons in these systems are strongly correlated with strong on-site interactions and thus the superconducting order parameters are considered to be necessarily anisotropic.

In connection with this problem it was pointed out that the antiferromagnetic spin fluctuations can mediate superconductivity of singlet pairing with d-wave character. From experimental investigations of the temperature dependence of the nuclear spin-lattice relaxation rate mainly d-wave character of the pairing symmetry of these systems was deduced. Importance of antiferromagnetic spin fluctuations in some of the heavy electron systems was recognized by observations of non-Fermi liquid properties around the antiferromagnetic QCP which was explicitly observed in many cases.

For the high-\( T_c \) cuprates we have quantitative information on the spectrum of spin fluctuations as explained in the preceding section. Thus the problem is to see if the high temperature superconductivity is explained with the use of the same spin fluctuation spectrum.

This problem was first treated by using a weak coupling (BCS) theory on a two dimensional square lattice Hubbard model consisting of \( d(x^2 - y^2) \) orbitals of Cu ions. Adjusting the relative size of the nearest and second nearest neighbor transfer matrices \( t_1 \) and \( t_2 \) as shown in Fig. 7 so as to reproduce observed Fermi lines, one can simulate the band structure of cuprates. Considering the spin fluctuation-mediated interaction between the electrons, the Hamiltonian is given by
\[ H = \sum_{\sigma k} \tilde{\varepsilon}_k \alpha^\dagger_{k\sigma} \alpha_{k\sigma} + \frac{1}{2N} \sum_{\sigma k} V_{kk'} \alpha^\dagger_{k\sigma} \alpha_{k'\sigma} \alpha^\dagger_{k'\sigma} \alpha_{k\sigma}, \]  
with
\[ V_{kk'} = \frac{3}{2} f^2[\chi(k-k') + \chi(k+k')], \]
where \( \tilde{\varepsilon}_k = \varepsilon_k - \mu \) is the single particle energy measured from the chemical potential. Using the BCS mean field approximation we have
\[ H \rightarrow \sum_{\alpha} \tilde{\varepsilon}_k \alpha^\dagger_{k\alpha} \alpha_{k\alpha} - \sum_{k} \{ \Delta(k) \alpha^\dagger_{k\uparrow} \alpha^\dagger_{-k\downarrow} + \text{h.c.} \} + \sum_{k} \Delta(k) \{ \alpha^\dagger_{k\downarrow} \alpha_{k\uparrow} \}, \]
with the gap function
\[ \Delta(k) = -\frac{1}{2} \sum_{k} V_{kk'} \langle \alpha^\dagger_{k\downarrow} \alpha^\dagger_{-k\uparrow} \rangle. \]

After diagonalizing [19] with the usual procedure we arrive at the following eigenvalue problem:
\[ \lambda \Delta(k) = \rho(k) \langle \mathcal{V}_{\mathcal{F}_\alpha} \Delta(k') \rangle_{\mathcal{F}_\alpha}, \]
where \( \rho(0) \) is the density of state at the Fermi level and the average \( \langle \cdot \rangle_{\mathcal{F}_\alpha} \) is taken over the Fermi surface. From the largest eigenvalue of \( \lambda \) the transition temperature is given by
\[ T_c = 1.13\omega_c e^{-1/\lambda}, \]
where \( \omega_c \) is the cutoff frequency. \( \chi(q) \) and \( \omega_c \) can be evaluated from the experimental results as discussed in the preceding section. Eq. [20] is solved numerically after symmetry classification of the solutions and the largest value of \( \lambda \) is obtained for the d-wave symmetry of \( (x^2 - y^2) \) type. The estimated value for \( T_c \) is of the order of 100 K, a result consistent with experiment. This result was reported in 1990\textsuperscript{19,38} and the d-\( (x^2 - y^2) \) type symmetry of the order parameter was confirmed experimentally after controversy of several years.

As a matter of fact the above estimated value of \( \lambda \) is near 1, indicating possible insufficiency of the weak coupling (BCS) theory. The strong coupling theory has been developed by using the dynamical susceptibility obtained from the SCR analysis of anomalous normal state properties.\textsuperscript{39} This approach deals with not only the pairing effects due to antiferromagnetic spin fluctuations but also depairing effects through scatterings of electrons by them. The normal and anomalous Green’s functions are defined by
\[ G(k, \tau - \tau') = -\langle T_\tau a^\dagger_{k\sigma}(\tau) a_{k\sigma}(\tau') \rangle, \]
\[ F'(k, \tau - \tau') = -\langle T_\tau a^\dagger_{k\sigma}(\tau) a^\dagger_{-k\sigma}(\tau') \rangle, \]
respectively, where \( \tau \) is the imaginary time and \( T_\tau \) is the imaginary time ordering operator. The anomalous Green’s function is nonzero in the superconducting state only. Considering the lowest order contribution of spin fluctuations to the self-energy (the Migdal approximation), the Dyson-Gor’kov equations for the Green’s functions are given as follows:
\[ [i\omega_n - \tilde{\varepsilon}_k - \Sigma^{(1)}(k,i\omega_n)]G(k,i\omega_n) - \Sigma^{(2)}(k,i\omega_n)F'(k,i\omega_n) = 1, \]
\[ [-i\omega_n - \tilde{\varepsilon}_k - \Sigma^{(1)}(-k,-i\omega_n)]F'(k,i\omega_n) + \Sigma^{(2)}(-k,-i\omega_n)^* G(k,i\omega_n) = 0, \]
with
\[ \Sigma^{(1)}(k,i\omega_n) = \frac{1}{N} \sum_{k'} \Sigma^{(1)}(k-k',i\omega_{n-m})G(k',i\omega_{n-m}), \]
\[ \Sigma^{(2)}(k,i\omega_n) = \frac{1}{N} \sum_{k'} \Sigma^{(2)}(k-k',i\omega_{n-m})F'(k',i\omega_{n-m}), \]
\[ V^{(q)}(q,i\omega_n) = i + F^2 \chi_s(q,i\omega_n) + (1)^{n-1} \chi_c(q,i\omega_n) - \frac{\chi_s(q,i\omega_m)}{\xi_s} + (1)^{n} \frac{\chi_c(q,i\omega_m)}{\xi_c}, (n = 1, 2), \]
where \( \chi_s(q,i\omega_n) \) and \( \chi_c(q,i\omega_n) \) are the spin and charge susceptibilities, respectively, \( \xi_s, \chi, \) etc., are the spin susceptibility for vanishing interaction, etc. and \( \omega_n = n\pi T \) is the Matsubara frequency. Near the antiferromagnetic instability the term with \( \chi_c \) is dominating and we can neglect the other terms. Thus we can calculate the self energy by using empirically determined values for \( \chi_s(q,i\omega_n) \) and \( \chi_c(q,i\omega_n) \). The dynamical susceptibility here is parameterized as in eq. [15, 16] with \( Q = (\pi, \pi), \theta = 0 \). The temperature dependence of \( \chi(Q) \) is calculated in terms of these parameters.

Now the equations [23, 24] are solved numerically for various models for the cuprates, i.e., for various band structures (transfer matrices), electron occupation, and coupling constant \( U \).\textsuperscript{40-43} We summarize here the main results.

(1) The superconducting order parameter has \( d - (x^2 - y^2) \) symmetry and the estimated values for \( T_c \) are consistent with the experimental results.

(2) According to systematic studies of the parameter dependence of \( T_c, T \), it depends most strongly on \( T_0 \) and is
approximately proportional to it. The $T_A$-dependence is less significant. The dependence on $y_0$ or the distance from the QCP is rather weak. The transfer parameter ratio ($t_2/t_1$) and the carrier concentration are not quite sensitive to the value of $T_c$ within reasonable ranges of their values.

(3) Two dimensional systems tend to have higher $T_c$ than three dimensional ones.\(^{44-46}\) When weak three dimensional character is introduced to two dimensional systems $T_c$ starts to decrease quite slowly, indicating that systems with layer structures are favorable for spin fluctuation-induced superconductivity.\(^{47}\)

(4) Experimental plots of $T_c$ against $T_0$ for high $T_c$ cuprates and heavy electron superconductors make a nearly straight line as shown in Fig. 8. This seems to indicate strongly that the cuprates and the heavy electron systems have the same origin of superconductivity. The points for the heavy electron superconductors are more scattered around the straight line compared with those for the high $T_c$ cuprates. This is considered to be natural since the formers have various different crystal structures.

The strong coupling theory was extended also to numerical works on the Hubbard and d-p models without having recourse to the experimental results, i.e., the dynamical susceptibility was calculated from the band structure. Although the Feynman diagrams for the SCR dynamical susceptibility were known, in practice the simplest approximation without vertex corrections has been employed widely. This is called the fluctuation exchange (FLEX) approximation and was derived from the Baym-Kadanoff formalism emphasizing the particle conservation.\(^{48}\) In what follows we briefly summarize the results of this approach.

(1) For the hole-doped systems the Hubbard and the d-p models were studied.\(^{49-54}\) In the latter model one considers $p\sigma$ orbitals of oxygen ions explicitly in addition to the $d(\chi^2-y^2)$ orbitals of copper. The energy difference of the $p$- and $d$-levels are determined consulting NMR and NQR experimental data. The values of the transfer integrals are chosen so as to reproduce well the observed Fermi lines. Choosing the value for $U/t$ within a reasonable range, superconductivity with $d(\chi^2-y^2)$ symmetry was obtained for a fairly large range of doping concentration. The values of $T_c$ were in a reasonable range. The following characteristic experimental results were explained with the FLEX calculations.

(a) The temperature variation of the order parameter near $T_c$ is much faster than in the BCS theory and the ratio $\Delta(0)/k_BT_c \approx 10$ is quite large compared with the BCS value 3.5. This is consistent with experiment.

(b) A sharp resonance peak at 41 meV was observed in neutron scattering measurements in the superconducting state of $YBa_2Cu_3O_{6+x}$. FLEX calculations of the dynamical susceptibility below $T_c$ showed a resonance peak around $(\pi, \pi)$ and was interpreted as the exciton or the bound state of an electron and a hole excited across the superconducting gap.

(c) Angle-resolved photoemission experiments show a peak-dip-hump structure of the one electron spectrum in the superconducting state. This result is well reproduced in the FLEX calculations.

(2) The electron-doped systems were studied with the Hubbard model and the experimentally observed differences between the electron- and hole-doped systems were well explained.\(^{55,56}\)

(a) The doping concentration range of antiferromagnetic phase is larger and that of the superconducting phase is smaller in the electron-doped systems than in the hole doped systems.

(b) The calculated value of $\Delta(0)/k_BT_c$ for the electron-
doped systems is half that for the hole-doped systems. This is just consistent with experimental results.

These differences between the electron- and hole-doped systems are considered to arise from the difference in the relative position of the hot spot (where the Fermi line crosses with the antiferromagnetic Brillouin zone boundary and the electrons are most strongly scattered by spin fluctuations) and the van Hove critical point where the density of states is large.

We show in Fig. 9 a phase diagram of quasi 2-dimensional systems calculated by the FLEX approximation in the temperature ($T$) against doping concentration ($\delta$) plane. Although the approximation is considered to be poor near $\delta = 0$ and realistic band parameters (transfer matrices) are different between the electron- and hole-doped systems this figure seems to reproduce the overall phase diagram of the doped cuprates fairly well (see Fig. 7). However, there is one discrepancy in the lower hole concentration side: The observed $T_c$ tends to decreases for small $|\delta|$ as $|\delta| \to 0$. This is related with the pseudo gap phenomena and will be discussed later.

It seems appropriate to discuss here very briefly on the superconductivity of 2-dimensional organic systems, $\kappa$-(BEDT-TTF)$_2$X. For example the compound with X = CuN(CN$_2$)Cl is an antiferromagnetic insulator at ambient pressure and under pressure a weak first order metal to insulator transition takes place at $p = 200$ bar. In the metallic phase the system shows superconductivity below $T_c = 13$ K. These systems are regarded as consisting of layers, each of which has an approximate square lattice of dimers of BEDT-TTF molecules. Relevant electrons in these systems are well described by a Hubbard model consisting of highest occupied molecular orbitals of dimers with just one hole per dimer. To a good approximation the electronic states are well simulated by choosing properly the transfer integrals shown in Fig. 10. This model is quite similar to that for high-$T_c$ cuprates. Furthermore, the superconducting phase is neighboring the antiferromagnetic phase in both systems. An important difference is that in the organic systems the band is just half-filled and the metallic state is clearly in the intermediate coupling regime, while in the cuprates the metallic state arises by doping an insulator.

The $t$-$J$ model approach from the strong coupling limit does not apply to the organic systems since it necessarily reduces to the insulating Heisenberg model for a system with a half-filled band. On the other hand the approach discussed here for the spin fluctuation mechanism well apply to both of the systems. The spin fluctuation-mediated superconductivity in these organic systems have been studied with the use of FLEX approximation. The results were the superconductivity with $d$-$(x^2-y^2)$ symmetry and the value of $T_c$ consistent with experiment. For further details we refer to 1).

It seems remarkable that the superconductivity in
the cuprates and that in 2-dimensional organic systems are consistently described in terms of the spin fluctuation mechanism within the same approximation. Furthermore, this seems to be the only available mechanism for the latter. In view of consistent descriptions for the high-$T_c$ cuprates, organic and heavy electron superconductors we may expect the spin fluctuation mechanism to be the common mechanism for all of these systems.

However, there are still unsolved problems associated with the pseudo gap phenomena observed in hole-underdoped cuprates and in $\kappa$-(BEDT-TTF)$_2$X salts. The pseudo gap region in the phase diagram of the cuprates may be seen in Fig. 5. These phenomena may be regarded as the low temperature corrections to the anomalous normal state properties discussed in the preceding section (see Fig. 6). For example, with decreasing temperature $1/T_1$ first increases following the CW law and then start to decrease in this region before reaching the superconducting transition temperature $T_c$ as if there is a spin excitation gap. Corresponding anomalies are observed in the resistivity, Hall coefficient and one-electron spectral density where pseudogaps are observed by angle-resolved photomission experiment. The $T_c$ against $\delta$ curve has a dome shape as in Fig. 5; $T_c$ decreases for small $|\delta|$ with decreasing $|\delta|$ as the antiferromagnetic phase is approached.

Although some part of the pseudogap phenomena including those observed in photoemission spectra may be explained concerning strong antiferromagnetic correlations in 2-dimensional systems, we clearly need additional mechanisms to explain all the other phenomena including the pseudogap as observed in the nuclear spin-lattice relaxation rate. One promising proposal may be to consider superconducting pairing fluctuations in addition to the spin fluctuations. Studies using FLEX for spin fluctuations and a t-matrix approximation for pairing fluctuations succeeded in describing various pseudogap phenomena and the trend of decreasing $T_c$ with decreasing $|\delta|$.[59] However, this approach does not seem to cover the region around the SC-AF line starting from $\delta = 0$. The overall success of the theory in describing main properties of the high-$T_c$ cuprates seems to indicate general soundness of the approaches.

7. Discussion and conclusion. In this article we have reviewed the theories of spin fluctuations and spin fluctuation mediated superconductivity as approaches from the magnetic QCP. The Overall success of the theory in describing main properties of the high-$T_c$ cuprates seems to indicate general soundness of the approaches.

It seems worth while here to take a wider scope of looking at the entire problem. We show in Fig. 11 a sketch of a phase diagram for the nearly half-filled Hubbard model in a three dimensional parameter space $T-U/t-\delta$. Appropriate approaches are indicated for shaded areas. We have no satisfactory theory in the blank area where the Mott transition in the $\delta = 0$ plane is included. This phase diagram is the one before considering the possibility of superconducting pairings. The effects of impurity potentials associated with the doping are neglected. Thus a part of the AFM (antiferromagnetic metallic) phase in reality may be AFI (AF insulator) due to the Anderson localization of carriers.

According to the theories discussed in this article superconducting phase appears in a fairly wide region along the QC line starting from $\delta = 0$ (organic superconductors, though neighboring discontinuously on AFI phase). An approximate position of cuprates is indicated in the figure. Various experimental results indicate that
$U/t$ is not quite large and the higher order terms in the $t/U$ expansion are significant. The value of $U/t$ deduced from experimental results falls between ~6 and ~8. Thus we are in the intermediate coupling regime and the approaches around the QC line as discussed in this article may be appropriate.

As was mentioned already we have at present no really satisfactory theory in the unshaded region of Fig. 11 and a part of the pseudogap area seems to belong to this difficult region which needs further theoretical studies. It may also be worth while to note that from this point of view the approaches from the limit of large $U/t$ and small $|\delta|$, i.e. the approach from the opposite side of the above-discussed one, seems necessarily to deal with this difficult region before arriving at the optimal area of the high-$T_c$ cuprates. From a theoretical point of view it is highly desirable to develop a theory which deals with this difficult area connecting continuously between the QCP area and the antiferromagnetic insulator phase ($\delta = 0$).

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