強相関電子系に対する動的分子場と
スピン揺らぎの統一理論の開発

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はしがき

酸化物高温超伝導体，重い電子系，有機導体などの強相関電子系の電子状態を理論的に正しく記述することは，現代の物性物理学における最大の課題である。酸化物超伝導体と有機導体については，電子相関の程度は中程度であると考えられているが，4f 電子を含む重い電子系化合物の多くは，強相関であるといえる。

強相関電子系の電子状態を記述するには，ハートリー・フォック近似は無力であり，LDA 近似を用いたバンド計算は，本質的には一体近似であるために，基底状態の性質以外は正しく記述できない。

強相関の極限とは，別の見方をすれば，局所的なクーロン斥力エネルギーがバンド・エネルギーをはるかに上回る原子極限であり，強相関物質の記述には，少なくとも，バンド極限と原子極限の両方で正しい理論が必要である。これに答える理論が，**動的分子場理論 (Dynamical Mean-Field Theory(DMFT))**[1,2]であるが，この理論はスピン系における分子場近似と対応しており，両者とも，空間次元 d が無限大の仮想的な極限では，厳密に正しい理論となる。したがって，d=3 においては，1/d の高次項を無視するという意味において近似理論となるが，半数充填の Hubbard 模型における Mott－Hubbard 転移を記述できる，金属の場合に正しくフェルミ液体となるなど，これまでの強相関電子系の理論にはない優れた性質を持っており，定量的にもよいことが，量子モンテカルロ法，有限系の厳密対角化法などとの比較から明らかにされている。また，電子系に対する動的分子場理論の，ハートリー・フォック理論（電子系の分子場理論と呼ばれることもある）やスピン系における分子場理論との大きな違いは，局所的な多体効果をすべて取り込みうることである。これは，動的分子場理論によれば，格子上の電子多体系が，有効媒質中の 1 個の（多体効果を伴う）不純物問題に帰着されるためである。この不純物問題を正しく解けば，局所的な効果は揺らぎも含めてすべて取り込むことができる。

しかしながら，動的分子場理論は，1/d の高次項を無視しているために，空間的に広がった揺らぎを取り入れることができない。この欠点に対し，1/d の高次の効果を系統的に取り入れる試みもなされている。それはすぐに予想されるとおり，d=∞における 1 個の不純物のかわりに，不純物のクラスターを扱う方法である。実際のやり方には 2 通りあり，(1)実空間における不純物クラスター[3]，または，(2)波数空間におけるクラスター[4]，をそれぞれ導入する。後者の方法は，ブリルアン・ゾーン内の波数を少数の点で代表させるため，代表波数間の距離が大きく，実空間では短距離の相関（小さなクラスター）を扱っていることになる。したがってこれらの方法では，たとえば量子臨界点近傍で重要になる，長距離（小さな波数）の相関効果は取り込めないことになる。また，これらの理論は長時間の数値計算を必要とし，バンド計算と組み合わせて現実の物質に適用するには向かない。

一方，量子臨界点におけるさまざまな異常現象を記述する理論としては，守谷による**自己無撞着なスピン揺らぎの理論** [5]が知られている。この理論は，弱相関の電子系を念頭に，

RPA 近似を出発点として，スピンの揺らぎを取り込む。いくつかの変形版があるが，最近の現象論的な，しかし本質をついた定式化では，動的帯磁率に対する総和則を巧みに用いて，スピン揺らぎの効果を取り込んだ量子臨界点の理論を構築している。守谷らは，これを強相関の重い電子系にも拡張して定式化し[6]，実験の解析にも成功している。

本研究の目的は，上に述べた 2 つの方法，「動的分子場理論」と「スピン揺らぎの理論」を統合し，強相関電子系に対しても用いることのできる，しかも，できる限りミクロな基礎付けを持った，「**動的分子場理論を超えてスピン揺らぎの効果を含んだ理論**」を構築することにあった。その構築にあたって，我々は，有効 1 不純物問題の解法としては反復摂動法を用い，動的分子場理論の計算を行った上で動的な局所帯磁率を求め，それを用いて格子系の動的帯磁率を求めた。それに対しスピン揺らぎの理論を適用し，総和則によって一様帯磁率の温度依存性を決めなおしている。スピン揺らぎの理論において長波長近似を用いることにより理論が簡潔になっているが，量子臨界点近傍の性質の記述は，元のスピン揺らぎの理論と同様の精度で可能である。

問題点としては，スピンの揺らぎも含めた完全な自己無撞着にはなっていないこと，反復摂動論を用いているために，完全にミクロな定式化にはなっていないこと，があげられる。これらは，$d<\infty$に対するクラスター理論と比較しながら，今後改良しなければならない。

スピンの揺らぎが重要な役割を果たしているといわれている現象としては，**熱電効果**（ゼーベック効果，ペルチエ効果）があげられる。最近，強相関電子系における熱電効果が注目されており，強い多体効果に起因してエネルギーに大きく依存する散乱が，大きな熱電変換効率をもたらすとして期待されている。しかしながら，強相関系における熱電効果はこれまで詳しく調べられていない。そこで，本研究では，以前から我々が研究してきた近藤絶縁体，とくに YbB_{12} と FeSi の熱電能についての研究を行った。FeSi についてはバンド計算で求められている状態密度を用い，これまで用いてきた 2-バンド模型に，正孔がわずかに含まれていると仮定すると，実験をよく説明できることがわかった。YbB_{12} については，同様の計算では低温側をあわせると高温側で実験と合わなくなることがわかった。これは，YbB_{12} がより強相関であることによる。計算の改良について現在検討中である。これらの結果を，熱電変換研究会における招待講演として発表した。ただし，熱電能については，動的分子場理論により，局所的なスピンの揺らぎは取り込んで計算しているものの，大域的なスピン揺らぎの効果を取り込む理論計算はまだできておらず，今後の課題である。スピン揺らぎは低エネルギーにおける電子の散乱にデリケートな影響をもたらすため，その効果を考慮した理論が求められている。

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Correlation Effects in Multi-Band Hubbard Model and Anomalous Properties of FeSi

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The two-band Hubbard model with the density of states obtained from the band calculation is applied for FeSi, which is suggested to be a Kondo insulator or a correlated band insulator. Using this model, the correlation effects on FeSi are investigated in terms of the self-consistent second-order perturbation theory combined with the local approximation. The calculated optical conductivity spectrum reproduces the experiments by Damascelli *et al.* semiquantitatively and the specific heat explains the anomalous contribution at about 250 K observed in FeSi. Inclusion of the spin fluctuation and the extension to the case of strong correlation are also discussed.

KEYWORDS: two-band Hubbard model, FeSi, optical conductivity, specific heat

In the study of a specific material among the strongly correlated electron systems, the effect of the band structures often plays a crucial role when one compares a theoretical calculation to the experiments. Use of a simple theoretical model might not capture the salient features of the material. Development of a theoretical method that is capable of taking proper account of the realistic features of the material is necessary. We report our recent approach to the study of the anomalous properties of FeSi in such direction.

FeSi is well known for more than thirty years and a number of studies from various aspects have been done, stimulated by the fascinating physical properties. The early study by Jaccarino *et al.*[1] showed that the susceptibility is much enhanced over the value expected from the band paramagnetism at finite temperatures and has a broad peak at about 500 K. It was also reported that the specific heat seems to have an anomalous enhancement at about 250 K. These behaviors were explained by a band model with an energy gap, but unphysically narrow bands were necessary, so that this difficulty has attracted interests of many researchers. From the conductivity measurements, FeSi is an insulator at low temperatures but shows metallic behavior at room temperature. To explain these unusual properties of FeSi, several theoretical approaches have been proposed, but the most successful one is the spin fluctuation scenario by Takahashi and Moriya.[2] It explains the anomalous magnetic property of FeSi and their idea of the thermally induced magnetic moment was confirmed by the neutron scattering experiment.[3]

The recent optical studies,[4–7] however, revealed the unusual properties of FeSi again. Schlesinger *et al.* reported that the gap of about 60 meV (~700 K) opened at low temperatures is filled and almost closed at room temperature (about 250~300 K), which they attributed to the correlation effect. The following experiments also reported the evidence of the correlation effects at low temperatures.[8–11] In these contexts, Aeppli and Fisk[12] suggested that FeSi can be viewed as a Kondo insulator or a strongly correlated insulator.

Kondo insulators have been found in the f-electron systems and typical examples are YbB_{12}[13] and $Ce_3Bi_4Pt_3$[14] and so on. They have correlated f-bands and small energy gaps at low temperatures. Although there are many similarities among FeSi and these materials, the correlation in FeSi may not be so strong. However, the same physics can be recognized both in FeSi and Kondo insulators, if one reexamines the experimental data carefully. From this aspect, Fu and Doniach[15] proposed an extended Hubbard model with two mixed conduction bands, which is based on their band calculation[16] for FeSi, and confirmed the importance of the correlation effects in physical quantities. Their calculation, however, seems to include some errors about the treatment of the self-energies. Therefore, we reinvestigated this model carefully and calculated the correlation effects in more correct way,[17] and confirmed that the correlation effects do play important roles, but the shape of the spectrum in the optical conductivity did not coincide with the experimental data, because of the use of the too simple model Hamiltonian.

Therefore in the present report, we use an extended two-band Hubbard model with the density of states obtained from the band calculation, and attempt to explain the low temperature anomalies of FeSi observed in the optical conductivity[18] and the specific heat consistently.

The band calculations[19–23] for FeSi predict that the ground state is a band insulator and a recent calculation[23] reproduces the gap size close to the observed one. Therefore, we start from the band insulator model, which consists of two Hubbard bands for d-electrons as

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follows.

$$H = \sum_{ij\sigma}(t^1_{ij}c^\dagger_{i1\sigma}c_{j1\sigma} + t^2_{ij}c^\dagger_{2i\sigma}c_{2j\sigma}) + U\sum_i(n_{i1\uparrow}n_{i1\downarrow} + n_{i2\uparrow}n_{i2\downarrow}) + U_2\sum_i(n_{i1\uparrow}n_{i2\downarrow} + n_{i2\uparrow}n_{i1\downarrow}) + U_3\sum_i(n_{i1\uparrow}n_{i2\uparrow} + n_{i2\downarrow}n_{i1\downarrow}) - J\sum_i(c^\dagger_{i1\uparrow}c_{i1\downarrow}c^\dagger_{i2\downarrow}c_{i2\uparrow} + c^\dagger_{i2\uparrow}c_{i2\downarrow}c^\dagger_{i1\downarrow}c_{i1\uparrow}), \quad (1)$$

where the $c^\dagger_{ia\sigma}(c_{ia\sigma})$ creates (destroys) an electron on site i in band a =1, 2 with spin σ. The tight binding parameters t^a_{ij} should be fitted to the band calculation and U, U_2, U_3 and J denote the Coulomb and exchange interactions.

Since one can expect that the optical conductivity spectrum reflects the structure of the quasi-particle density of states (DOS) of a system, we use the DOS obtained from the band calculation for FeSi by Yamada *et al.*[23] for the initial DOS so as to enable detailed comparison with the experiment.

Furthermore, we start from the following general expression of the current operator,

$$j = e\sum_{\sigma,\mathbf{k}}\sum_{mm'} v^{mm'}_{\mathbf{k}} c^\dagger_{m\mathbf{k}}c_{m'\mathbf{k}}, \quad (2)$$

where m denotes the band indices and derive the convenient expression for the optical conductivity. For simplicity, we set the intra- and interband contributions to be equal ($v^{mm'}_{\mathbf{k}} = v_{\mathbf{k}}$). Moreover, we assume that the momentum conservation is violated in real systems by some defects and phonon-assisted transitions. Therefore, using the linear response theory, we consider the current-current correlation function as below,

$$K(i\omega_n) = \int_0^\beta d\tau e^{i\omega_n\tau}\sum_{mm'}\sum_{\mathbf{k}\mathbf{k}'\sigma\sigma'} v_{\mathbf{k}}v_{\mathbf{k}'} \times < T_\tau c^\dagger_{m\mathbf{k}\sigma}(\tau)c_{m\mathbf{k}\sigma}(\tau)c^\dagger_{m'\mathbf{k}'\sigma'}(0)c_{m'\mathbf{k}'\sigma'}(0) >$$

$$\simeq -\frac{1}{\beta}\sum_{mm'}\sum_{l}\sum_{\mathbf{k}\mathbf{k}'\sigma} v_{\mathbf{k}}v_{\mathbf{k}'}\mathcal{G}^m_{\mathbf{k}\sigma}(i\nu_l)\mathcal{G}^{m'}_{\mathbf{k}'\sigma}(i\nu_l + i\omega_n) \times[\delta_{\mathbf{k}\mathbf{k}'} + \Gamma^{mm'\sigma}_{\mathbf{k}\mathbf{k}'}(i\nu_l; i\omega_n)\mathcal{G}^m_{\mathbf{k}'\sigma}(i\nu_l)\mathcal{G}^{m'}_{\mathbf{k}\sigma}(i\nu_l + i\omega_n)], \quad (3)$$

where $\Gamma^{mm'\sigma}_{\mathbf{k}\mathbf{k}'}(i\nu_l; i\omega_n)$ denotes the vertex function, and set [...] constant. For the present case, this leads to the following expression for the optical conductivity,

$$\sigma(\omega, T) = \frac{\pi(ev)^2}{\hbar}\sum_\sigma\int_{-\infty}^{\infty} d\nu\frac{f(\nu) - f(\nu+\omega)}{\omega} \times[\rho^\sigma_1(\nu) + \rho^\sigma_2(\nu)][\rho^\sigma_1(\nu+\omega) + \rho^\sigma_2(\nu+\omega)], \quad (4)$$

where $\rho^\sigma_a(\nu)$ denotes the DOS for the band a. This joint-DOS-like form for the optical conductivity is simple but convenient for the present case. We set $(ev)^2/\hbar = 1$ for simplicity.

Firstly, we show the optical conductivity obtained from the Hartree-Fock approximation (HFA) or a rigid band model in Fig. 1. The used DOS is displayed in Fig. 2 by the solid line for $T = 0$. The DOS is independent of the temperature within HFA. At 0 K, only the interband contribution survives and reproduces the shape of the spectrum of the experiment at 4 K in Fig. 3. Therefore, the band calculation by Yamada *et al.*[23] seems to give a good result about the whole structure of the DOS at $T = 0$ but with a slightly smaller gap size (see the comparison with the experiment below). Within the rigid band model, however, since the gap is filled only with the intraband (Drude) contribution, the temperature variation is monotonous and the spectrum does not become flat at a temperature of the order of the gap size. This disagreement was shown by Fu *et al.* first. Ohta *et al.*[5] also calculated the optical conductivity in the joint-DOS form from their band calculation, but the flat part of the optical conductivity spectrum within the gap could not be reproduced. Therefore, the rigid band model is not sufficient to explain the experiments.

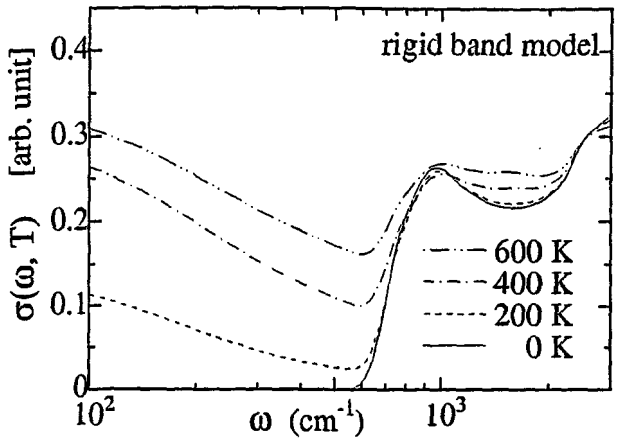

Fig. 1. Fig. 1 Calculated optical conductivity within the Hartree-Fock approximation or a rigid band model.

Next, we investigate the correlation effect in the low energy and low temperature region of this model. Therefore we calculate the correlation effect by the self-consistent second-order perturbation theory (SCSOPT) combined with the local approximation. The second-order self-energies are given by

$$\Sigma^{(2)\sigma}_1(\omega) = \iiint_{-\infty}^{\infty} d\varepsilon_1 d\varepsilon_2 d\varepsilon_3 [U^2\rho^{-\sigma}_1(\varepsilon_1)\rho^\sigma_1(\varepsilon_2)\rho^{-\sigma}_1(\varepsilon_3) + U_2^2\rho^{-\sigma}_2(\varepsilon_1)\rho^\sigma_1(\varepsilon_2)\rho^{-\sigma}_2(\varepsilon_3) + U_3^2\rho^\sigma_2(\varepsilon_1)\rho^\sigma_1(\varepsilon_2)\rho^\sigma_2(\varepsilon_3) + J^2\rho^{-\sigma}_2(\varepsilon_1)\rho^\sigma_2(\varepsilon_2)\rho^{-\sigma}_1(\varepsilon_3)] \times\frac{f(-\varepsilon_1)f(\varepsilon_2)f(\varepsilon_3) + f(\varepsilon_1)f(-\varepsilon_2)f(-\varepsilon_3)}{\omega + \varepsilon_1 - \varepsilon_2 - \varepsilon_3 + i\delta},$$

$$\Sigma^{(2)\sigma}_2(\omega) = (1 \leftrightarrow 2), \quad (5)$$

where $\rho^\sigma_a(\omega) = -(1/\pi)\mathrm{Im}G^\sigma_a(\omega + i\delta)$ and

$$G^\sigma_a(\omega) = \frac{1}{N}\sum_{\mathbf{k}} G^\sigma_a(\mathbf{k}, \omega)$$

$$= \int_{-\infty}^{\infty} d\varepsilon \rho_a^{0\sigma}(\varepsilon) \frac{1}{\omega - \varepsilon - \Sigma_a^{(2)\sigma}(\omega)}. \quad (6)$$

Here, N is the number of sites, $f(\varepsilon)$ the Fermi function and $\rho_a^{0\sigma}(\varepsilon)$ the DOS of band a for the non-interacting case. To make numerical calculation easy, we take δ finite ($\delta = 10^{-7}$) in eq. (6) and convert these equations with the transformations[25]

$$A_a^\sigma(\tau) = \int_{-\infty}^{\infty} d\epsilon e^{-i\tau\epsilon} \rho_a^\sigma(\varepsilon) f(\varepsilon),$$
$$B_a^\sigma(\tau) = \int_{-\infty}^{\infty} d\epsilon e^{-i\tau\epsilon} \rho_a^\sigma(\varepsilon) f(-\varepsilon). \quad (7)$$

These equations have to be solved self-consistently. In this paper, we set $U_2 = U - J$ and $U_3 = U - 2J$ in order to reduce the number of parameters. In this case, the Hamiltonian is rotationally invariant in spin and real spaces if the two bands are degenerate.[24]

In the following results, $U = 0.5$ eV and $J = 0.35U$ are chosen so as to reproduce the shape and the temperature dependence of the optical conductivity spectrum. The solid line for $T = 0$ in Fig. 2 indicates the initial DOS at 0 K, and the correlation effect is absent except the Hartree-Fock contribution since the band 1 is filled and the band 2 is empty.

Note that the gap in the DOS is widened by 16 % so as to reproduce the shape of the spectrum of the optical conductivity at 4 K in the experiment, which does not change the essence of the following result. Then, the gap size (E_g) of 75 meV is obtained if the steepest parts of the DOS at the both sides of the gap are extrapolated and the tails are neglected. (If we regard the gap as the region inside the tails of the gap edge, we obtain 60 meV.) The band 1 and 2 in our Hamiltonian correspond to the upper and lower part of the DOS with respect to the Fermi level ($E_F = 0$) as is seen in Fig. 2, where we introduce a cut off for each band so as to include one state per spin in each band. Then the band width for the band 1 and 2 are about 0.56 eV and about 0.85 eV, respectively. Although the DOS is asymmetric, the chemical potential is fixed at $\omega = 0$ and assumed to be temperature independent. One can see in Fig. 2 that the correlation is introduced at finite T through the thermally excited electrons and holes and the gap existing at 0 K is almost filled up at the temperature of the order of its size, which results in the temperature variation of the interband contribution of the optical conductivity (see below).

In Fig. 3(a), the temperature variation of the optical conductivity calculated from the temperature-dependent DOS in Fig. 2 is shown. In our calculation (Fig. 3(a)), the gap is almost filled up at 300 K as well as the rapid increase in the gap region from 150 to 300 K is seen. This is consistent with the experiment (Fig. 3(b)), where the gap is filled rapidly from 100 K to 300 K. Reflecting the correlation effects, the peak at the gap edge shifts to lower frequency region, as is seen in the experiment. In our calculation, however, there are dips between the Drude and the interband contributions in contrast to the experiment. This may be caused by the simplification in deriving eq. (4). However, the almost flat spectrum is obtained at 300 K, which comes from the temperature

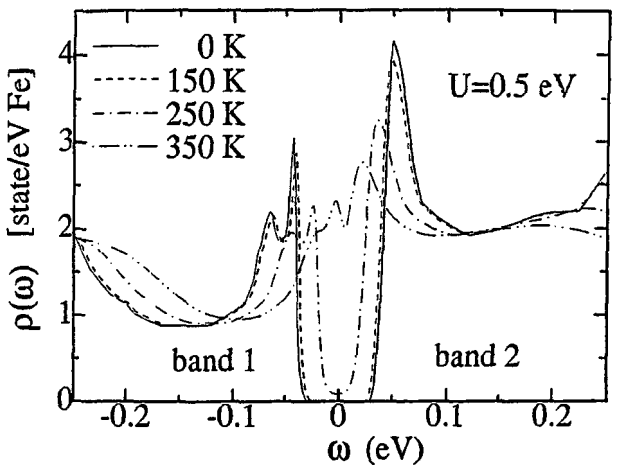

Fig. 2. The temperature dependence of the quasi-particle DOS and the initial DOS obtained from the band calculation(Ref. 26) at $T = 0$. At finite T, the DOS is strongly temperature dependent due to the correlation effects.

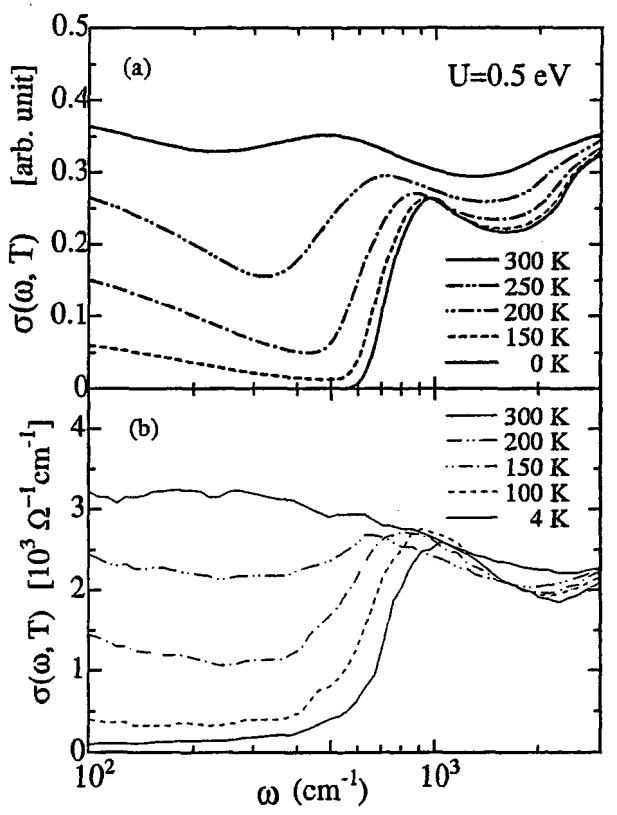

Fig. 3. (a)The temperature dependence of the optical conductivity calculated with the eq. (4). (b)The experimental data from Ref. 11. The peaks due to phonons observed in the gap are omitted.

dependence of the interband contribution.

We also calculate the temperature variation of the specific heat with the same parameters as in the optical conductivity. Starting from the equation of motion,[26] we obtain the following expression for the total energy per

site:

$$E = \frac{1}{2}\sum_{\sigma}\int_{-\infty}^{\infty} d\omega f(\omega)\left[\omega\{\rho_1^{\sigma}(\omega)+\rho_2^{\sigma}(\omega)\}\right.$$
$$\left.+\frac{1}{N}\sum_{\mathbf{k}}\{\varepsilon_{\mathbf{k}}^1\rho_1^{\sigma}(\mathbf{k},\omega)+\varepsilon_{\mathbf{k}}^2\rho_2^{\sigma}(\mathbf{k},\omega)\}\right], \quad (8)$$

where $\varepsilon_{\mathbf{k}}^1$ ($\varepsilon_{\mathbf{k}}^2$) is the Fourier transformation of t_{ij}^1 (t_{ij}^2).

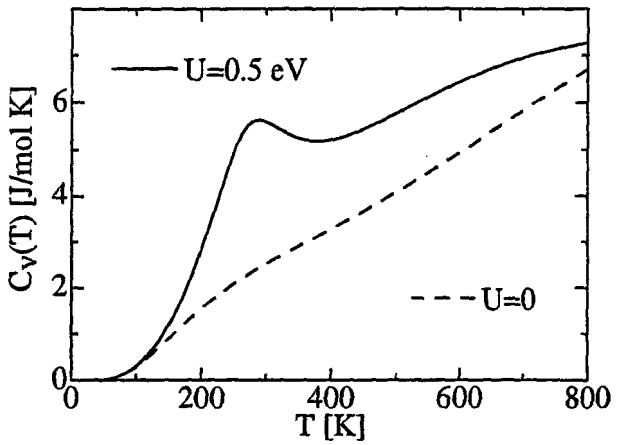

Fig. 4. Calculated specific heat using the same parameters as in Fig. 3(a).

The specific heat can be calculated from the numerical differentiation of the energy as $C_V = (\partial E/\partial T)_V$. The difference between the cases with $U = 0$ and 0.5 eV in Fig. 4 indicates the contribution from the correlation effect, which results in a peak of about 4 J/K mol at about 250 K, and explains the "anomalous" contribution (~6 J/K mol) in the specific heat at about 250 K reported by Jaccarino *et al.*[1] Note that they evaluated the anomaly by subtracting the specific heat of CoSi after the normal electronic contributions γ_{FeSi} and γ_{CoSi} are removed, respectively. In the above calculations, we confirmed that the correlation effect is essential to explain the temperature dependence of the optical conductivity and the specific heat in FeSi. At higher temperatures or for magnetic properties, however, it is also important to take the spin fluctuations[2,27] into account.

The self-consistent renormalization (SCR) theory of spin fluctuations has succeeded in describing the itinerant magnetism and the quantum critical phenomena with a small number of parameters.[28] On the other hand, the dynamical mean field theory (DMFT) is one of the most powerful schemes to take account of the strong local correlation. One of the authors has proposed a new and practical scheme that unifies DMFT and SCR in a microscopic way.[27] Application of this theory to FeSi may improve the present calculation towards the inclusion of the effects of spin fluctuations at finite temperatures and the intermediate coupling.

Acknowledgements

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Hybrid theory of the dynamical mean field and the spin-fluctuations in strongly correlated electron systems

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Abstract

In order to take account of the spatial fluctuation beyond the dynamical mean field theory (DMFT), the two-particle self-consistent theory is carefully reinvestigated and is proved to be unable to reproduce the correct low energy scale in the strong correlation limit. An improved theory is proposed which starts from DMFT and is combined with the spin-fluctuation theory (SFT) in a phenomenological form. The present theory therefore encompasses DMFT and SFT, and describes the quantum critical behavior properly with the same exponent as SFT. The local quantum dynamics is fully taken into account as in DMFT in contrast to the phenomenological treatment in SFT.

Key words: dynamical mean field theory, two-particle self-consistent theory, spin-fluctuation theory, strongly correlated electron systems

The most powerful method for the description of the strongly correlated electron systems (SCES) may be the dynamical mean-field theory (DMFT).[1] It, however, lacks the effect of intersite spin fluctuations, which becomes important, e.g. in the vicinity of the quantum phase transitions.[2] Several methods were proposed to overcome this deficiency of DMFT by taking account of the cluster instead of the effective impurity in DMFT,[3,4] but none of them has succeeded in the description of the quantum critical phenomena (QCP) because of the use of the finite size of the cluster. The extended DMFT is proposed by Si and Smith[5] for the fermion-boson model, which can describe the properties at QCP. The other successful approachs to QCP are the spin fluctuation theories (SFT's).[6,2] The SFT by Moriya was further developped

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in a form which is more phenomenological but flexible in use.[9] The two-particle self-consistent theory (TPSC)[7] has been proposed to improve SFT. We, however, recently proved that TPSC cannot reproduce the correct low energy scale in the strong correlation limit of the single impurity Anderson model (SIAM), and therefore cannot be a good candidate for an improved theory.[8] The proof proceeds as follows. For the symmetric case of SIAM, the TPSC equation for the spin sector

$$T\sum_{\omega}\frac{2\Pi_0(\mathrm{i}\omega)}{1-U_s\Pi_0(\mathrm{i}\omega)}=n-2\langle n_\uparrow n_\downarrow\rangle, \tag{1}$$

reads as

$$1-\frac{U_s}{2U}\simeq\frac{2}{\pi^2}\log\frac{\omega_c}{\Delta-U_s/\pi}, \tag{2}$$

where ω_c denotes a cutoff frequency of the order $O(\Delta)$ (resonance width), and we have used the low energy form for the poralization function $\Pi_0(\omega)$. The above equation determines the effective interaction U_s in the spin channel and the static susceptibility $\chi_s = 1/2(\pi\Delta - U_s) = 1/4T_K$. However, the above equation (2) yields the Kondo temperature $T_\mathrm{K} \to (\pi/2)\omega_c \exp(-\pi^2/2)$ for $U \to \infty$ which is finite and does not vanish.

We then propose a theory which avoids the above defect and combines both DMFT and SFT. This is done first by introducing the effective vertex $\Gamma(\omega) = U/[1 + U\Pi_0(\omega)]$ and expressing the local dynamical susceptibility approximately as ($g\mu_B = 1$)

$$\chi_L(\omega)=\frac{\frac{1}{2}\Pi_0(\omega)}{1-\Gamma(\omega)\Pi_0(\omega)}, \tag{3}$$

where $\Pi_0(\mathrm{i}\omega) = -T\sum_\epsilon G(\mathrm{i}\epsilon)G(\mathrm{i}\epsilon + \mathrm{i}\omega)$ and the Green's function $G(\mathrm{i}\epsilon)$ is the solution of DMFT. $\chi_L(0)$ diverges when and only when $U \to \infty$ at $T = 0$, which is the desired property for impurity. Actually, we modify this as

$$\tilde{\chi}_L(\omega)=\frac{1}{\chi_L(\omega)^{-1}-\mathrm{i}C\omega}, \tag{4}$$

and determine the parameter C by the sum rule, eq.(1), to correct the low energy scale as a fine tuning.

Next task is to construct the wave-vector-dependent dynamical susceptibility $\chi(q,\omega)$ by taking account of the $1/d$ corrections. In $d \to \infty$ limit, $\chi(q,\omega)$ can

be calculated from the knowledge of the effective impurity[1] as

$$\chi(q, \mathrm{i}\omega) = \sum_{\epsilon\epsilon'} \left[\frac{\chi_q^0(\mathrm{i}\omega)}{1 - \Gamma(\mathrm{i}\omega)\chi_q^0(\mathrm{i}\omega)} \right]_{\epsilon\epsilon'}, \tag{5}$$

where

$$\chi_q^0(\mathrm{i}\omega)_{\epsilon\epsilon'} = -\sum_k G(k, \mathrm{i}\epsilon) G(k+q, \mathrm{i}\epsilon' + \mathrm{i}\omega)\delta_{\epsilon\epsilon'}, \tag{6}$$

and $\Gamma(\mathrm{i}\omega)_{\epsilon\epsilon'} = \Gamma(\mathrm{i}\epsilon, \mathrm{i}\epsilon', \mathrm{i}\omega) = [\chi_L^0(\mathrm{i}\omega)^{-1} - \chi_L(\mathrm{i}\omega)^{-1}]_{\epsilon\epsilon'}$ is the vertex function which is local and the matrix of the Matsubara frequencies ϵ, ϵ' and $\chi_L(\mathrm{i}\omega) = \sum_{\epsilon\epsilon'} \chi_L(\mathrm{i}\omega)_{\epsilon\epsilon'}$. $G(k, \mathrm{i}\epsilon)$ is the Green's function in $d \to \infty$.

For $d < \infty$, the calculation of $\chi(q, \omega)$ is not easy, but it has a general form as

$$\chi(q, \omega) = [\chi_{\mathrm{L}}(\omega)^{-1} - J(q, \omega)]^{-1}. \tag{7}$$

Therefore, we rather adopt Moriya's approach[9] and use the approximate long-wavelength expansion form around the ordering vector Q:

$$\chi(Q + q, \omega) = [\tilde{\chi}_{\mathrm{L}}(\omega)^{-1} - J_Q(T) + Aq^2]^{-1}, \tag{8}$$

and determin $J_Q(T)$ by the sum rule similar to [1] but including the wave-vector sum. If $J_Q(T = 0)$ is chosen so that $\chi(Q, \omega = 0, T = 0)^{-1} = \tilde{\chi}_L(\omega = 0, T = 0)^{-1} - J_Q(T = 0) = 0$ (QCP), the specific heat, staggered susceptibility and resistivity show $T^{3/2}$ behaviors at low temperatures[10] in accord with the SFT.[9] An example for the Hubbard model[10] is shown in Fig. 1.

The present theory thus encompasses DMFT[1] and SFT[9], and describes the quantum critical behavior properly with the same exponent as SFT.[8] The local quantum dynamics is fully taken into account as in DMFT in contrast to the phenomenological treatment in SFT. It could acquire a microscopic basis if one would be able to calculate $J(q, \omega)$ microscopically up to $O(1/d)$. Furthermore, a formula for calculating the self-energy from $\chi(q, \omega)$ is not yet established in the case of strong correlation except for the fermion-boson model.[12] For $d \to \infty$, the self-energy can be calculated with a rather good accuracy by using the iterative perturbation theory[11]. An extension of it to the case with orbital degeneracy would be a future issue[13].

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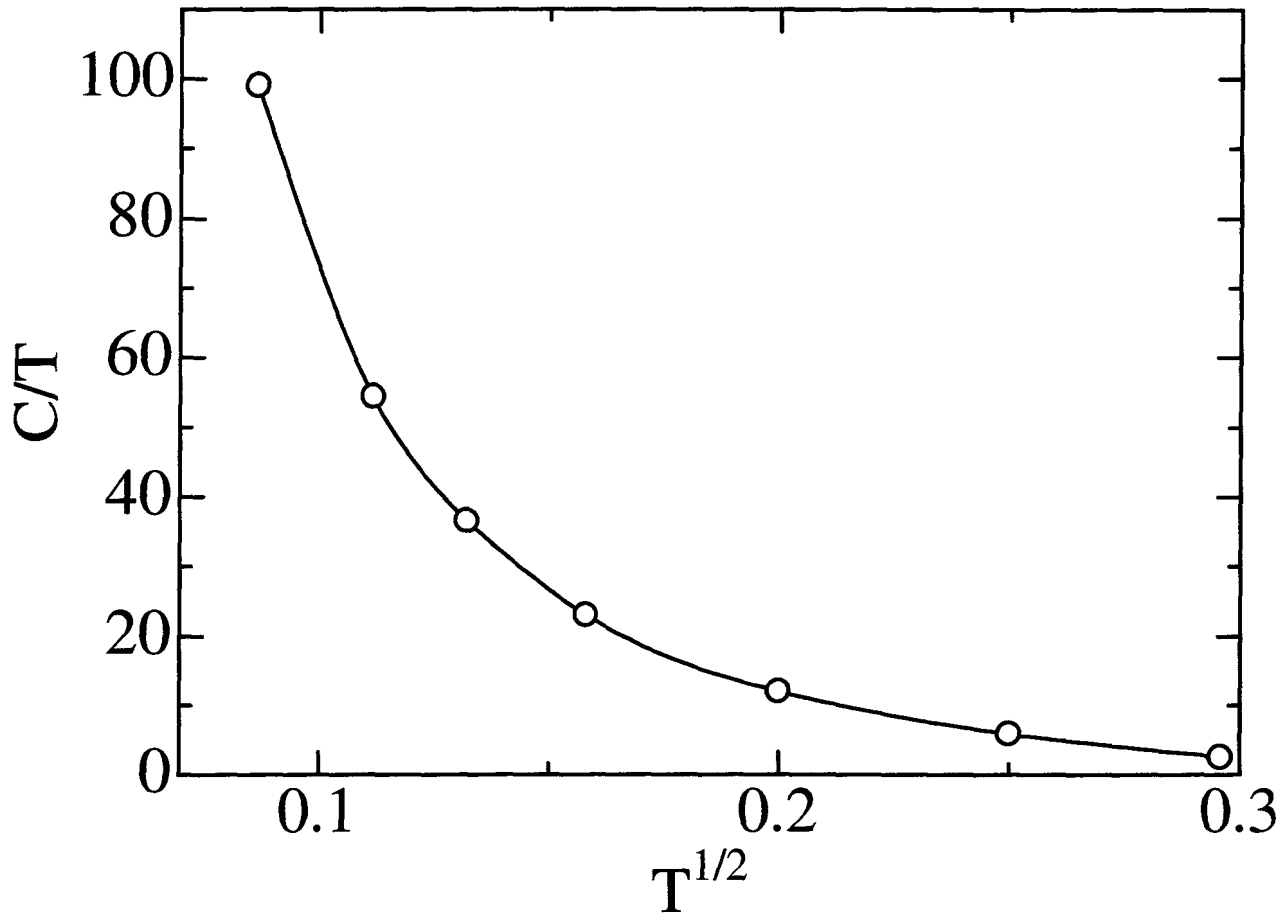

Fig. 1. Temperature dependence of $C(T)/T$ of the Hubbard model at QCP is plotted.

Spin Fluctuation Theory for Heavy Fermions and Kondo Insulators

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Spin fluctuation theory for strongly correlated electron systems is constructed. First, the two-particle-self-consistent theory is applied to the single-impurity Anderson model. It is found that it cannot reproduce the small energy scale in the strong correlation limit. A modified scheme to overcome this difficulty is proposed by introducing an appropriate vertex correction explicitly. Using the same vertex correction, the self-energy is investigated, and it is found that under certain assumptions it reproduces the result of the modified perturbation theory which interpolates the weak and the strong correlation limits. Based on these, a new spin fluctuation theory which is applicable to the case of strong correlation is proposed.

KEYWORDS: spin fluctuation, two-particle self-consistent theory, dynamical susceptibility, self-energy

§1. Introduction

The dynamical mean field theory[1] (DMFT) is the most powerful method for describing the effect of strong correlation. It becomes exact in the limit of large spatial dimension $d \to \infty$, where the lattice problem is reduced to solving an impurity problem embedded in an effective medium self-consistently. Thus the local fluctuation is fully taken into account, but the spatially extended fluctuations are neglected. In order to describe the spatial fluctuations, the so-called self-consistent renormalization (SCR) theory[2] was proposed, first for d-electron systems, where the correlation is not very strong, and it was extended later to the case of strong correlation phenomenologically.[3] SCR theory describes a system by only small number of parameters, and hence details of the structure specific to the system are not taken into account. Purpose of the present paper is to propose a theory for strongly correlated systems which starts from DMFT and interpolates between DMFT and SCR.

For this purpose, we first investigate the two-particle self-consistent theory (TPSC)[4] and apply it to the single-impurity Anderson model (SIAM) in §2. TPSC can be regarded as an extension of SCR theory for weak correlation,[2] and has proved to be successful in the study of the Hubbard model in the weak and the intermediate correlations. It is considered to be superior to the fluctuation-exchange approximation (FLEX)[5] in that only TPSC can reproduce the side peaks in the single-particle spectrum in the case of strong correlation.

We will, however, show in §2 that it cannot reproduce a small energy scale in SIAM in the strong correlation limit.[6] In §3 we propose an improved scheme which is applicable to the case of strong correlation by introducing vertex corrections. We also investigate the self-energy using the same approximation and find that under certain assumptions our theory reproduces the modified perturbation theory (MPT),[7–9] which interpolates the weak and the strong correlation limits. Thus the phenomenological character of MPT is partially resolved. Finally, in §4 we construct a theory for the lattice system with strong correlation which interpolates DMFT and SCR theories.[10] It is applicable to heavy electron materials and Kondo insulators. The effect of spin fluctuation leads to the quantum critical behavior at the phase boundary, in which the critical exponents are the same as those by SCR but the properties specific to the materials can be taken into account beyond the ordinary SCR theory.

§2. The Two-Particle-Self-Consistent Theory for the Single-Impurity Anderson Model

The Hamiltonian for SIAM is written as

$$\mathcal{H} = \sum_{k\sigma} \epsilon_k c^+_{k\sigma} c_{k\sigma} + E_f \sum_{\sigma} n_{f\sigma} + V \sum_{k\sigma} (f^+_\sigma c_{k\sigma} + c^+_{k\sigma} f_\sigma) + U n_{f\uparrow} n_{f\downarrow} \quad (2.1)$$

in the ordinary notation, where ϵ_k and E_f denote the energy of the conduction and f electrons, respectively, V the hybridization, and U the Coulomb repulsion between f-electrons. In the following, the subscript f in $n_{f\sigma}$ will be dropped. The orbital degeneracy is neglected.

In TPSC, the charge and the spin susceptibilities are expressed in the form similar to those of RPA, but with the renormalized interaction parameter U_c and U_s for charge and spin channels, respectively. These parameters are determined by the sum rules,

$$T \sum_{\omega} \frac{2\Pi_0(i\omega)}{1 + U_c \Pi_0(i\omega)} = n + 2\langle n_\uparrow n_\downarrow \rangle - n^2, \quad (2.2)$$

$$T \sum_{\omega} \frac{2\Pi_0(i\omega)}{1 - U_s \Pi_0(i\omega)} = n - 2\langle n_\uparrow n_\downarrow \rangle. \quad (2.3)$$

Here ω denotes the Matsubara frequency, $n = \langle n_\uparrow \rangle + \langle n_\downarrow \rangle$ and U_s are related to $\langle n_\uparrow n_\downarrow \rangle$ by $U_s = U\langle n_\uparrow n_\downarrow \rangle / \langle n_\uparrow \rangle \langle n_\downarrow \rangle$. Thus, U_s can be determined from the second equation, while U_c can be obtained by solving the first equation once U_s is solved. If we set $g\mu_B = 1$ ($g = 2$ is the

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g-factor), the static susceptibility $\chi \equiv \chi^{zz}$ is given by

$$\chi(\omega=0) = \frac{1}{2}\frac{\Pi_0(0)}{1-U_s\Pi_0(0)} = \frac{1}{2\pi\Gamma}, \tag{2.4}$$

where $\Pi_0(0) = 1/\pi\Delta$. It is well known that χ is proportional to the inverse of the Kondo temperature T_K and diverges to infinity for $U \to \infty$ in the symmetric case since T_K vanishes. I have proved,[6] however, that it is not the case. I obtained $T_K \to (\pi/2)\omega_c \exp(-\pi^2/2)$ for $U \to \infty$ (ω_c is a cutoff) which is finite. Therefore, it is clalified that TPSC can not be applied to the strong correlation limit.

§3. Improvement of Two-Particle-Self-Consistent Theory

The dynamical susceptibility of the Anderson impurity can be generally expressed by the diagram shown in Fig.1 and the equation,

$$\chi^{+-}(i\omega) \equiv T\sum_{\epsilon\epsilon'} \chi^{+-}(i\epsilon, i\epsilon', i\omega), \tag{3.1}$$

$$\chi^{+-}(i\epsilon, i\epsilon', i\omega) = -G_\uparrow(i\epsilon)G_\downarrow(i\epsilon'+i\omega) \times \left[\delta_{\epsilon,\epsilon'} + T\sum_{\epsilon''}\Gamma(i\epsilon'', i\epsilon', i\omega)\chi^{+-}(i\epsilon'', i\epsilon', i\omega)\right]. \tag{3.2}$$

We calculate the irreducible vertex function $\Gamma(i\epsilon, i\epsilon', i\omega)$ approximately by taking account of the maximally crossed diagrams (Fig.2) as

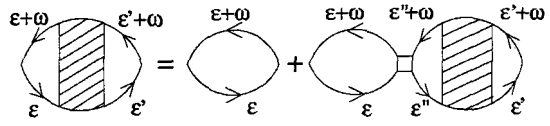

Fig. 1. The Feynman diagram for the susceptibility $\chi^{+-}(i\epsilon, i\epsilon', i\omega)$. The square denotes the vertex function $\Gamma(i\epsilon, i\epsilon', i\omega)$.

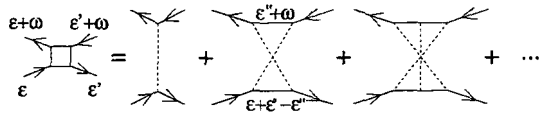

Fig. 2. The diagram for the vertex function $\Gamma(i\epsilon, i\epsilon', i\omega)$.

$$\Gamma(i\epsilon, i\epsilon', i\omega) = \frac{U}{1+UK(i\epsilon+i\epsilon', i\omega)}, \tag{3.3}$$

$$K(i\epsilon+i\epsilon', i\omega) = T\sum_{\epsilon''} G_\downarrow(i\epsilon+i\epsilon'-i\epsilon'')G_\uparrow(i\epsilon''+i\omega). \tag{3.4}$$

Here the unperturbed Green's function $G_{0\sigma}$ should be substituted for G_σ, and K is denoted as K_0. Then, we find that $K_0(0,\omega) = K_0(\omega,0) = \Pi_0(\omega)$. ($\Pi_0(\omega)$ denotes the polarization function.) If we approximate $\Gamma(i\epsilon, i\epsilon', i\omega)$ by $\Gamma(i\omega) \equiv \Gamma(0,0,i\omega)$, then we obtain

$$\chi^{+-}(i\omega) = \frac{\Pi_0(i\omega)}{1-\Gamma(i\omega)\Pi_0(i\omega)}, \tag{3.5}$$

$$\Gamma(i\omega) = \frac{U}{1+U\Pi_0(i\omega)}. \tag{3.6}$$

Note that at $T = 0$ and the low frequency limit, the effective interaction becomes

$$\Gamma(0) = \frac{U}{1+U/\pi\Delta}, \tag{3.7}$$

which smoothly interpolates between $\Gamma(0) = U$ for small U and $\Gamma(0) \to \pi\Delta$ for $U \to \infty$. (Δ denotes the resonance width of the f level.) Therefore, the magnetic susceptibility $\chi^{zz}(0) = (1/2)\chi^{+-}(0)$ diverges when and only when $U \to \infty$. This is a desired property. Within the present approximation, eq.(3.5) becomes

$$\chi^{+-}(i\omega) = \Pi_0(i\omega)[1+U\Pi_0(i\omega)]. \tag{3.8}$$

Namely, the terms higher than the second order with respect to U in the perturbational expansion of $\chi(\omega)$ are completely canceled out with the terms from the vertex correction, which is of course an artifact of the approximation. Note also that eq.(3.8) is correct up to $O(U)$.[10,11]

Actually, eq.(3.8) does not satisfy the sum rule eq.(2.3), so that we previously proposed[10] to modify $\chi^{+-}(\omega)$ into

$$\chi^{+-}(\omega)' = \frac{1}{\chi^{+-}(\omega)^{-1} - iC\omega}, \tag{3.9}$$

and determine the parameter C by the sum rule. This is essentially corresponding to modifying the low energy scale. The imaginary parts of the dynamical susceptibility calculated with this C term correction are shown in Fig.3. We set $\Delta = 1$ here and henceforth.

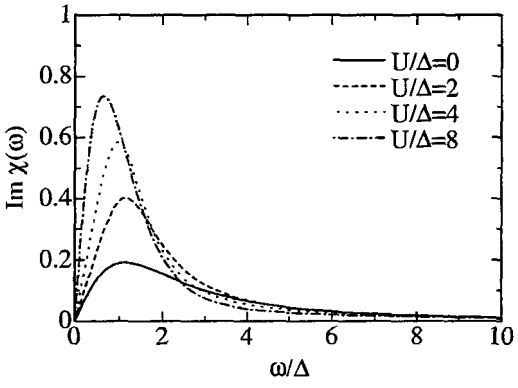

Fig. 3. The imaginary parts of the dynamical susceptibility $\chi^{+-}(\omega)'$ are shown for U/Δ =0, 2, 4 and 8.

Next, we investigate the self-energy. The Feynman diagram is shown in Fig.4, which may be expressed as

$$\Sigma_\uparrow(i\epsilon) = Un_\downarrow + \cdots$$

$$+T^3\sum_{\epsilon',\epsilon'',\omega,\cdots}\Gamma(i\epsilon,i\epsilon',i\omega)G_\uparrow(i\epsilon')G_\downarrow(i\epsilon'+i\omega)$$
$$\times\Gamma(i\epsilon',i\epsilon'',i\omega)\cdots G_\uparrow(i\epsilon'')G_\downarrow(i\epsilon''+i\omega)$$
$$\times\Gamma(i\epsilon'',i\epsilon,i\omega)\times G_\downarrow(i\epsilon+i\omega)+\cdots. \quad (3.10)$$

Keeping the behaviors of $K_0(0,\omega)$ and $K_0(\epsilon,0)$ in mind,

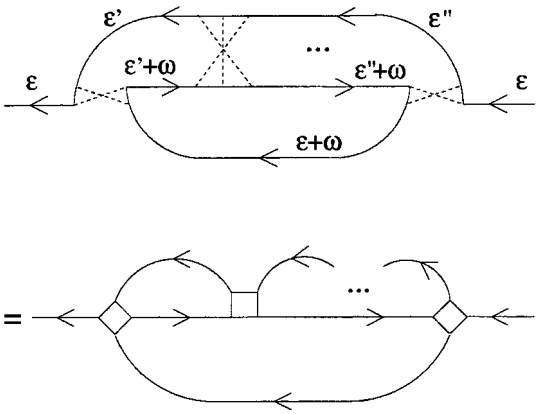

Fig. 4. A typical Feynman diagram for the self-energy.

we approximate $\Gamma(i\epsilon,i\epsilon',i\omega)$ at the left and the right ends by the separable form $[\bar{\Gamma}(i\epsilon)\Gamma(i\omega)]^{1/2}$, where $\bar{\Gamma}(i\epsilon)$ denotes some average of $\Gamma(i\epsilon,i\epsilon',i\omega)$ on ϵ' and ω. $\Gamma(i\omega)$ is given by eq.(3.6) and $\bar{\Gamma}(i\epsilon)$ is approximated as

$$\bar{\Gamma}(i\epsilon)\simeq\langle\Gamma(i\epsilon,i\epsilon,i\omega)\rangle_\omega\simeq\frac{U}{1-B'\Sigma^{(2)}_\uparrow(i\epsilon)/Un_\downarrow}. \quad (3.11)$$

where B' is an adjustable parameter. This form is inferred by comparing the first- and second-order self-energies.[6] Now the self-energy is written as

$$\Sigma_\uparrow(i\epsilon)\simeq Un_\downarrow+$$
$$\bar{\Gamma}(i\epsilon)T\sum_\omega\Gamma(i\omega)\frac{\Pi_0(i\omega)}{1-\Gamma(i\omega)\Pi_0(i\omega)}G_{0\downarrow}(i\epsilon+i\omega). \quad (3.12)$$

We further note that

$$\frac{\Gamma(i\omega)}{1-\Gamma(i\omega)\Pi_0(i\omega)}=U, \quad (3.13)$$

which holds within the present approximation. Therefore we obtain

$$\Sigma_\uparrow(i\epsilon)=Un_\downarrow+\frac{U^2T\sum_\omega\Pi_0(i\omega)G_{0\downarrow}(i\epsilon+i\omega)}{1-B'\Sigma^{(2)}_\uparrow(i\epsilon)/Un_\downarrow}$$
$$=Un_\downarrow+\frac{\Sigma^{(2)}_\uparrow(i\epsilon)}{1-B\Sigma^{(2)}_\uparrow(i\epsilon)}, \quad (3.14)$$

where $B=B'/Un_\downarrow$. This is exactly the same form as the interpolative self-energy proposed by Martin-Rodero, et al.,[7] which bridges between the second-order and the atomic limit self-energies in both the symmetric and the asymmetric cases. B was determined so as to reproduce the correct self-energy in the atomic limit when $U\to\infty$. Namely, one obtains $B=(1-2n_{-\sigma})/Un_{-\sigma}(1-n_{-\sigma})$.

Since the above formula for the self-energy does not satisfy the Friedel sum rule, one has to introduce an effective d level energy to be adjusted to fulfil the sum rule,[8,9] or to subtract $\Sigma(0)$ from $\Sigma(\epsilon)$[11] to approximately satisfy it. Details of the calculations will be found in ref.9. Note that B vanishes for the symmetric case. Namely, the self-energy becomes equal to the second-order one due to the cancelation of the higher order terms in the present approximation.

§4. Spin Fluctuation Theory for the Lattice

In this section, we first discuss the dynamical susceptibility of strongly correlated electron systems (SCES). In extending SCR theory to the case of SCES, Moriya and Takimoto[3] assumed the following form for the dynamical susceptibility, motivated by the duality picture of Kuramoto and Miyake:[12]

$$\chi(Q+q,\omega)=[\chi_{\rm L}(\omega)^{-1}-J_Q(T)+Aq^2]^{-1}, \quad (4.1)$$

where $\chi_{\rm L}(\omega)$ denotes the local susceptibility of each magnetic ion and Q an ordering vector. In SCR, $\chi_{\rm L}(\omega)^{-1}$ is expanded up to the linear term of ω, yielding

$$\chi^{\rm SCR}_{\rm L}(\omega)=\frac{\chi_L}{1-i\omega/\Gamma_{\rm L}}, \quad (4.2)$$

below certain cutoff frequency ω_c. Here, $\Gamma_{\rm L}$ is of the order of the Kondo temperature of the magnetic ion. In contrast to SCR theory, we retain full dynamical structure of $\chi_{\rm L}(\omega)$.

In DMFT, the dynamical susceptibility $\chi(q,\omega)$ of the lattice can be obtained once the effective impurity problem was solved.[1] Namely,

$$\chi(q,i\omega)=\sum_{\epsilon\epsilon'}\left[\frac{\chi^0_q(i\omega)}{1-\Gamma(i\omega)\chi^0_q(i\omega)}\right]_{\epsilon\epsilon'}, \quad (4.3)$$

where

$$\chi^0_q(i\omega)_{\epsilon\epsilon'}=-\sum_k G(k,i\epsilon)G(k+q,i\epsilon'+i\omega)\delta_{\epsilon\epsilon'}, \quad (4.4)$$

and $\Gamma(i\omega)_{\epsilon\epsilon'}=\Gamma(i\epsilon,i\epsilon',i\omega)$ is the vertex function. The procedure, however, needs rather tedious calculations. Ohkawa[13] and Miyake and Narikiyo[14] discussed a general form of $\chi(q,\omega)$ and derived the following equation

$$\chi(q,\omega)=[\chi_{\rm L}(\omega)^{-1}-J(q,\omega)]^{-1} \quad (4.5)$$

where $J(q,\omega)\simeq U^2\Delta\chi_0(q,\omega)+\lambda(q,\omega)$ in the strong correlation limit. $\Delta\chi_0(q,\omega)$ denotes the intersite part of the dynamical susceptibility of the lattice,

$$\chi_0(q,i\omega)=-T\sum_{\epsilon\epsilon'}\chi^0_q(i\omega)_{\epsilon\epsilon'}, \quad (4.6)$$

$$\Delta\chi_0(q,i\omega)=\chi_0(q,i\omega)-\frac{1}{N}\sum_q\chi_0(q,i\omega), \quad (4.7)$$

and $\lambda(q,\omega)$ contribution from the mode-mode coupling of spin fluctuations. $J(q,\omega)$ includes both the mean field contribution to the exchange interaction, which is $O(1/d^0)$, and the fluctuations from the mean field, which is $O(1/d)$. Here we assume that $J(q,\omega)$ can be expanded around a certain ordering vector Q as $J(Q+q,\omega)=J_Q(T)-Aq^2+\cdots$ and ω-dependence can be neglected.

Then we obtain $\chi(Q+q,\omega)$ in the same form as eq.(4.1). We can determine $J_Q(T)$ by the condition that the spin-fluctuation amplitude

$$\bar{S}_{\rm L}^2 = \frac{3}{\pi}\int_0^\infty d\omega[1+2n(\omega)]{\rm Im}\chi_Q(\omega) \qquad (4.8)$$

stays constant: $\bar{S}_{\rm L}^2(T_N) = \bar{S}_{\rm L}^2(T)$, where T_N is the ordering temperature. Both quantum and thermal fluctuations are included in $\bar{S}_{\rm L}^2(T)$. When there is no long range order, we set $T_N = 0$ in the above condition. In eq.(4.8), $\chi_Q(\omega)$ is defined by

$$\chi_Q(\omega) = \frac{1}{N}\sum_q \chi(Q+q,\omega). \qquad (4.9)$$

The frequency dependence of $\chi_Q(\omega)$ and the temperature dependence of $\chi(Q,0)$ are plotted in Figs.5 and 6 for J_Q =0.5, 0.6, 0.7 and J_c. It is seen that the low energy structure of $\chi_Q(\omega)$ becomes singular and much narrower than $\chi_{\rm L}(\omega)$, and $\chi(Q,0)$ becomes proportional to $T^{3/2}$ at QCP, as expected.[3)]

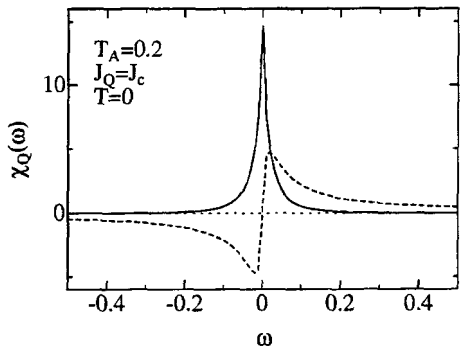

Fig. 5. The real (solid line) and imaginary (broken line) parts of the dynamical susceptibility $\chi_Q(\omega)$ at $T = 0$ are plotted.

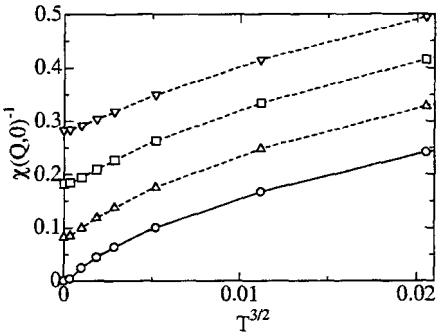

Fig. 6. The susceptibility $\chi_Q(0)$ is plotted as a function of T for J_Q =0.5, 0.6, 0.7 and J_c from upper to lower curves.

Once these calculations are done, we recalculate the self-energy. It must be done in a manner consistent with the IPT calculation. We proposed to use the formula

$$\Sigma(i\epsilon) = U^2 T\sum_\omega \tilde{G}(i\epsilon+i\omega)\tilde{\chi}_Q(i\omega), \qquad (4.10)$$

$$\tilde{\chi}_Q(i\omega) = \tilde{\Pi}(i\omega) + \frac{1}{N}\sum_q \frac{J(Q+q,i\omega)\tilde{\Pi}^2(i\omega)}{1-J(Q+q,i\omega)\chi_{\rm L}(i\omega)} \qquad (4.11)$$

where $J(q,\omega)$ is replaced with $J_Q(T) - Aq^2$. Note that the second term of $\tilde{\chi}_Q(\omega)$ has the same criticality as $\chi_Q(\omega)$. If we set $J(Q+q,i\omega) = 0$ in this form, the self-energy eq.(4.10) recovers the IPT result.

It is seen that ${\rm Im}\Sigma(\epsilon)$ at $\epsilon \sim 0$ becomes singular in contrast to the Fermi liquid behavior ${\rm Im}\Sigma(\epsilon) \propto \epsilon^2$ in IPT. Theoretical analysis leads to ${\rm Im}\Sigma(\epsilon) \propto \epsilon^{3/2}$ at QCP and $T = 0$.

Using the above self-energy, we calculate the density of states again. We find that the peak at E_F becomes thinner and singular due to the strong renormalization by the spin fluctuation.

Finally, the total energy is calculated from

$$E = 2\int_{-\infty}^{\infty} d\epsilon f(\epsilon)\left(-\frac{1}{\pi}\right){\rm Im}[\{\epsilon - \frac{1}{2}(Un_{-\sigma} + \Sigma(\epsilon))\}G(\epsilon)]. \qquad (4.12)$$

The specific heat is obtained by numerical derivative of E as $C = \partial E/\partial T$. We find that $C(T)/T \propto -\sqrt{T}$ at QCP as in SCR.[3)]

The electrical resistivity in the dimensionless form is calculated by the formula,[3)]

$$R(T) = \int_0^\infty d\omega\, n(\omega)[1+n(\omega)]{\rm Im}\chi_Q(\omega). \qquad (4.13)$$

Since this formula is an approximate one, we do not take care whether we should use $\tilde{\chi}_Q(\omega)$ in stead of $\chi_Q(\omega)$. Furthermore, the behaviors of $\chi_Q(\omega)$ and $\tilde{\chi}_Q(\omega)$ are the same at the critical point. The numerical result seems consistent with the theoretical analysis $R(T) \propto T^{3/2}$.[3)]

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Seebeck Coefficient of Kondo Insulators

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Seebeck coefficient S of the Kondo insulators is investigated theoretically within the framework of the dynamical mean field theory. It is found that the temperature dependence changes from the ordinary behavior $S(T) \propto T^{-1}$ in semiconductors to approximately $S \propto T$ at low temperatures due to the finite imaginary part of the electron self-energy in the Kondo insulators with strong correlation. Realistic models for YbB_{12} and FeSi based on the band calculations are also investigated.

KEYWORDS: Seebeck coefficient, thermoelectric power, Kondo insulators, strong correlation

§1. Introduction

Thermoelectric power (TEP) is currently attracting renewed interest because of the increasing need for a portable refrigerator without use of vapor coolant.[1] Narrow gap semiconductors have been intensively studied so far for such purpose and Bi_2Te_3 and its alloys are found to be the most efficient material (highest figure of merit $Z = S^2/\kappa\rho$, where S, κ and ρ denote, respectively, Seebeck coefficient, the thermal couductivity and the resistivity). It has been known that the magnetic alloys with the Kondo impurities exhibit large TEP. This is because of the strong energy-dependence of the carrier relaxation time due to Kondo-type scattering in these materials, and has been studied theoretically by using the non-crossing approximation and the Bethe Ansatz approach.[2-4] The latter yields

$$S(T) \cong \frac{\gamma}{e} \cot\left(\frac{\pi n_f}{N}\right) T, \tag{1.1}$$

at low temperatures, where γ, n_f and N denote the specific heat coefficient, f-electron number and the degeneracy of f-orbit, respectively. Thus the formula predicts the positive (negative) for Ce (Yb) ions, mostly in agreement with the experiments. The TEP makes a peak around the Kondo temperature T_K and the absolute value decreases above it.

The many heavy fermion materials also show large TEP ($|S| \lesssim 100$ μV/K) and can be understood by the same mechanism as the Kondo alloys mentioned above. In some of the compounds (e.g. $CeCu_2Si_2$), however, the TEP changes the sign at low temperatures, which can not be explained by the above mechanism. This phenomena is sometimes attributed to the effect of magnetic fluctuations, but a details are still not clear.

Recently, however, materials with much higher values of $|S|$ were found in the compounds called Kondo insulators. For example, YbB_{12}, a typical Kondo insulator with the energy gap of the order of 100K, shows $|S|_{\max} \sim 140$ μV/K at arount T =100 K,[15] and FeSi, the Kondo insulator of the transition metal element, exhibited $S_{\max} \sim 500$ μV/K at T =50 K.[6] These materials show large TEP only at rather low temperatures, but it may be advantageous for a refrigerator which works at low T.

It is well known[1] that $S(T) \sim -(k_B/e)\ (E_{c,v} - \mu)/k_BT$ for semiconductors, where $E_{c,v}$ denotes the gap edge position of the conduction or valence bands and μ the chemical potential. Our first concern in the present study is how this is modified by the strong correlation in the Kondo insulators. We study it by using the dynamical mean-field theory (DMFT)[7] for the periodic Anderson model (PAM), and will find that the rise of S in proportion to T^{-1} turns into approximately $S \propto T$ at low temperatures due to the finite imaginary part of the electron self-energy. We also study two-band models with the density of states calculated from the APW band calculations and apply them to YbB_{12} and FeSi. Comparisons with experiments and discussions will be presented in the last section.

§2. Boltzmann Equation Approach

Seebeck coefficient S is given by the sum of the electron diffusion and the phonon drag terms. As will be found below, the former is enhanced in most of the strongly correlated materials. Therefore, we focus only on the electron term and call it simply as S. By using the Boltzmann equation, S is given by

$$S(T) = -\frac{1}{eT} \int d\epsilon L(\epsilon)(\epsilon - \mu) \left(-\frac{\partial f}{\partial \epsilon}\right) \Big/ \int d\epsilon L(\epsilon) \left(-\frac{\partial f}{\partial \epsilon}\right), \tag{2.1}$$

where $L(\epsilon) \equiv \rho_c(\epsilon) v_c(\epsilon)^2 \tau_c(\epsilon)$ and $\rho_c(\epsilon)$, $v_c(\epsilon)$ and $\tau_c(\epsilon)$ denote the density of states (DOS), velocity and the relaxation time of conduction electrons, respectively. This formula yields the well-known result $S(T) \sim -(k_B/e)\ (E_{c,v} - \mu)/k_BT$ for semiconductors. On the other hand, the Peltier coefficient Π is related to S as $\Pi = TS$, hence $\Pi \sim (E_{c,v} - \mu)/e$ remains finite even at low temperature limit. The Peltier coefficient is defined as the heat absorbed or emitted at the junction of two elements when a unit charge flows through it. Therefore, it would turn out that the third law of the thermodynamics is bro-

ken if Π remains finite at $T \to 0$. Careful investigation of the electron-phonon coupled Boltzmann equation, taking full account of the nonequilibrium state of the phonon system, did not resolve this contradiction.[8] In the present paper, we will show below that the many-body effect will resolve this problem through the self-energy which is not included in the Boltzmann approach.

§3. Effect of Strong Correlation

In DMFT,[7] the TEP is given by the same formula as (2.1) but with

$$L(\epsilon) = \frac{1}{\pi N} \sum_{\mathbf{k}} v_{c\mathbf{k}}^2 [\mathrm{Im} G_c(\boldsymbol{k}, \epsilon)]^2 , \qquad (3.1)$$

where $G_c(\boldsymbol{k}, \epsilon)$ and $v_{c\mathbf{k}}$ are the Green's function and the velocity of conduction electrons. The vertex correction drops out in this theory.

Schweitzer and Czycholl[9] applied this scheme to the PAM and calculated the TEP for the metallic cases by using the self-consistent second-order perturbation theory. We here investigate the case of the Kondo insulators.

The Kondo insulators are the band insulators with strong correlation between f (or d in the case of FeSi) electrons.[10] The PAM is the simplest model for them. We use the DMFT scheme mentioned above and calculate the self-energy by the iterative perturbation theory in a modified form (mIPT).[11]

To evaluate eq.(3.1), however, one has to perform the k-summation over the Brillouin zone, which requires tedious numerical calculations. When the anisotropy is absent and the damping of quasi-particle is weak, however, $L(\epsilon)$ can be approximated by $L(\epsilon) \simeq v_F^2 \rho_c(\epsilon) \tau_c(\epsilon)$. Here the velocity is assumed constant and replaced by the Fermi velocity v_F. In the case of the PAM, $\rho_c(\epsilon)$ and $\tau_c(\epsilon)$ are given by

$$\rho_c(\epsilon) = -\frac{1}{\pi N} \mathrm{Im} \sum_k \frac{1}{\epsilon - \epsilon_k - \dfrac{V^2}{\epsilon - E_f - \Sigma_f(\epsilon)}}, \qquad (3.2)$$

$$\tau_c(\epsilon)^{-1} = -2\mathrm{Im} \frac{V^2}{\epsilon - E_f - \Sigma_f(\epsilon)}, \qquad (3.3)$$

where E_f and $\Sigma_f(\epsilon)$ denote the level position and the self-energy of an f-electron. V denotes the mixing and ϵ_k the energy of the conduction electrons.

The results are shown in Fig. 1 for the various f-electron level positions. The DOS of the conduction band is assumed to be a semicircular form with the half-width $W = 1$ around $\epsilon = 0$ and the resonance width of the f-level is set $\Delta = 0.5$. The Coulomb repulsion U between f-electrons is chosen as $U = 2$. $E_f = -1$ corresponds to the so-called symmetric case and express the Kondo insulator, and the other cases with $E_f > -0.7$ are metallic and hole-like, while $E_f = -0.7$ and -0.8 are insulators. $E_f = -0.6$ is marginal. These may corrspond to the Ce compounds. The calculations with $E_f < -1$ yield the same results with the opposite sign of S and may correspond to the Yb compounds. In the present model, the TEP can be finite only when there is an electron-hole asymmetry. The cases $E_f = -0.7$ and -0.8 show steep rises of S at low temperatures similar to the ordinary semiconductors as mentioned in §2, but S turns to decrease almost linearly at lower temperatures. This is because the quasi-particle DOS $\rho_c(\epsilon)$, which has an energy gap of the order of the Kondo temperature at $T = 0$, becomes temperature-dependent, so that the DOS within the gap becomes finite at finite temperatures. Note that the inverse relaxation time τ_c^{-1} near E_F decreases to very small values at low temperatures also, but the product $\rho_c(\epsilon)\tau_c(\epsilon) \propto L(\epsilon)$ exhibit a gap at $T = 0$. This gap is filled up gradually at finite temperatures. However, the numerical calculation at the lowest temperatures is delicate, and we could not determine the value of $L'(0)/L(0)$ at low temperature limit precisely, so that $S(T) \propto T$ is obtained only approximately.

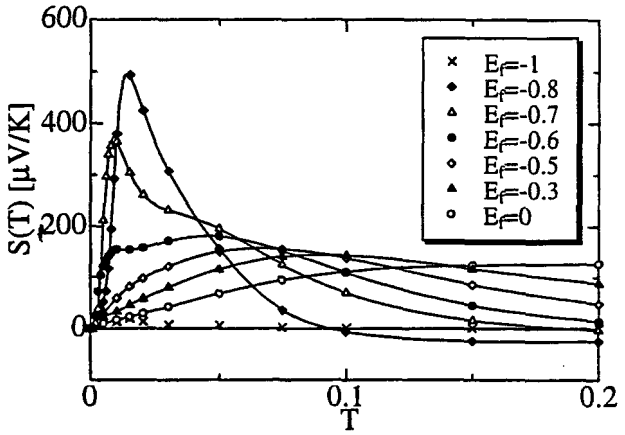

Fig. 1. The thermoelectric power of the periodic Anderson model for various values of E_f.

§4. Two-band Models for FeSi and YbB_{12}

In order to understand the behavior of TEP for a specific material, it is important to start from the knowledge of a band calculation since the TEP is sensitive to the details of the electronic structure. For most of the Kondo insulators, the band calculations exhibit energy gaps at the Fermi levels. The simplest way to express these results is to use a two-band model, each of which is expressed by the DOS of the corresponding bands located below and above E_F obtained from the band calculation. The mixing between f and conduction electrons is already included in these bands. Therefore, the Coulomb repulsion could have band-off-diagonal terms, even if it were diagonal in orbitals in the form of PAM. But since we do not have such knowledge at hand, we simply introduce the Coulomb interactions only within the same band and neglect the interband terms. Also, we neglect the effect of f-level degeneracy in YbB_{12} and treat only the spin degeneracy. A full analysis including the orbital degeneracy is to be done in the next stage.

When we analyze the experimental data, we should not forget the effect of the nonstoichiometry or impurities. Both shift the chemical potential into the conduction or valence bands, so that the material behaves as a metal at lowest temperatures. The experimental data for TEP of FeSi and YbB_{12} show $S(T) \propto T$ at low temperatures. A

careful experiment will clarify whether these are due to the nonstoichiometry/impurities or the many-body effect mentioned in §2. In FeSi, it was found that the electron-doping by Ir leads to negative S.[6]

First we show the results for FeSi, using the DOS calculated by Yamada.[12] Assuming the 0.01% hole-doping, we have obtained a good agreement with the experiment as shown in Fig. 2. Introduction of an intermediate value of $U = 0.5$ eV, which gives rise to reasonable agreement with the experiment on the dynamical conductivity and the specific heat,[13] changes the result only slightly since the many-body effect becomes effective only at temperatures higher than 100K in this material.[13]

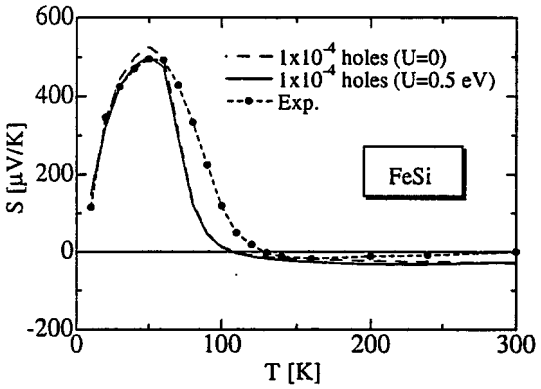

Fig. 2. The thermoelectric power of FeSi for hole-doped case with and without Coulomb interaction is compared with the experiment.

Fig. 3 shows the results for YbB_{12} calculated without the correlation effect. The DOS is taken from the LDA+U band calculation by Harima.[14] The peak at 10K in the experimental curve[15] is considered to be due to the phonon-drag effect. The calculation for stoichiometric case ($n = 0$) shows a diverging upturn at lowest temperature and does not fit the experiment. The curves with 0.5% or 1% electron-doping seem to be consistent with the experiment as regards the second peak at 40K, which may be due to the f-electrons. However, the high temperature behavior is not consistent with the experiment. In YbB_{12}, the many-body effect is considered to be important at low temperatures, so that the density of state may be strongly renormalized and becomes temperature-dependent. The effective f-electron position may be shifted from the vicinity to the Fermi level at low temperatures to deeper positions at higher temperatures than T_K, so that S becomes more electron-like and negative at high temperatures, improving the agreement with the experiment. Such a calculation including the correlatin effect is now in progress.

§5. Discussions

The present study has investigated the Seebeck coefficient of the Kondo insulators in terms of the dynamical mean-field theory. The vertex corrections and the Umklapp processes are not included in this formulation. Effects of these have been studied for high T_c cuprates within the framework of the fluctuation-exchange (FLEX) theory and were found to be significant.[18,19] However,

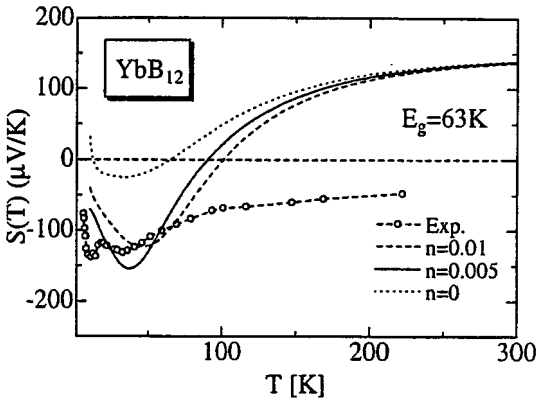

Fig. 3. The thermoelectric power of the periodic Anderson model for various values of E_f.

such effects are considered to be not not profound when the anisotropy is weak in the three-dimensional materials. The effect would be mainly to renormalize the absolute value of the conductivity, but may not affect the Seebeck coefficient seriously since it is expressed by the ratio of the transport integrals as in eq.(2.1).

Origins of the T-linear TEP at low temperatures in FeSi and YbB_{12} are still not clarified. Whether it is due to the nonstoichiometry or the many-body effect must be uncovered by careful experimental and theoretical studies in the future.

Acknowledgements

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Transport properties through a quantum dot in Kondo regime by modified perturbation theory

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Abstract Using the modified perturbation theory for the impurity Anderson model, we calculate the transport properties through a quantum dot in Kondo regime.[1] The method is based on the second order perturbation theory with respect to the Coulomb repulsion, but we modify the self-energy so as to reproduce the correct atomic limit and to fulfil the Friedel sum rule exactly. In the electron-hole symmetric case, when odd number of electrons exist in the dot, the Kondo peak appears at zero bias in the differential conductance (dI/dV) as is observed in recent experiments.[2, 3, 4] In the electron-hole asymmetric case, different ways of applying bias voltage, which is given by the difference of the chemical potentials between the left and right leads as $V=\mu_L-\mu_R$, have different effects on the shape of the dI/dV.

It has been pointed out that the Kondo effect in a quantum dot system has brought up new and interesting issues for physics, *e.g.*, the tunable Kondo effect or the nonequilibrium Kondo effect. Several theoretical methods have been devised to explain Kondo-type transport through a quantum dot using the impurity Anderson model.[5, 6, 7, 8, 9, 10]

The Hamiltonian for a quantum dot connected to the leads is written as

$$H = \sum_{\nu,k,\sigma} \varepsilon_k^\nu n_{k,\sigma}^\nu + \sum_\sigma \varepsilon_0 n_\sigma + U n_\uparrow n_\downarrow + \sum_{\nu,k,\sigma} V_k^\nu (c_{k,\sigma}^{\nu\dagger} c_{0,\sigma} + c_{0,\sigma}^\dagger c_{k,\sigma}^\nu), \quad (1)$$

where ε_0 and U represent an energy level and the Coulomb repulsion in a dot. The change of ε_0 is equivalent to the change of the gate voltage in measurements. ε_k^ν denotes the conduction electron energy in the lead $\nu(=R$ and $L)$. V_k^ν denotes the coupling between leads and a dot. We neglect orbital degeneracy and the k dependence of V_k^ν.

The equilibrium Green's function for the electron in the dot is given by

$$G_\sigma(\omega) = (\omega - \varepsilon_0 - U n_{-\sigma} - \Sigma_\sigma(\omega) + \mathrm{i}\Gamma)^{-1}, \quad (2)$$

where $\Sigma_\sigma(\omega)$ is the self-energy to be calculated and $\Gamma/2 = \Gamma_L = \Gamma_R = \pi\rho_c(0)V^2$ is the resonant level width and $\rho_c(0)$ denotes the density of states (DOS) of conduction electrons at the Fermi level. In our modified perturbation theory, we introduce the effective energy level $\tilde{\varepsilon}_0$ in place of the Hartree-Fock level $\varepsilon_0 + U n_{-\sigma}$, where n_σ denotes the electron number, and determine it so as to satisfy the Friedel sum rule exactly and to reproduce correct atomic-limit simultaneously. Thus the effective first-order Green's function is given by

$$G_\sigma^{(1)}(\omega) = (\omega - \tilde{\varepsilon}_{0\sigma} + \mathrm{i}\Gamma)^{-1}. \quad (3)$$

Using this Green's function, we first calculate the ordinary second-order self energy $\Sigma_\sigma^{(2)}(\omega)$. Then, we introduce the modified self-energy which is correct in the atomic limit as follows:

$$\Sigma_\sigma(\omega) = \Sigma_\sigma^{(2)}(\omega)/\{1 - B\Sigma_\sigma^{(2)}(\omega)\}, \quad (4)$$

where $B = \{U(1-n_{-\sigma}^{(1)}) + \varepsilon_0 - \tilde{\varepsilon}_{0\sigma}\}/\{U^2 n_{-\sigma}^{(1)}(1-n_{-\sigma}^{(1)})\}$, $n_\sigma^{(1)} = n[G_\sigma^{(1)}]$ and $n[G_\sigma] \equiv \int d\omega f(\omega)\ (-1/\pi)\mathrm{Im}G_\sigma(\omega)$. Next, we construct the second-order Green's function as

$$G_\sigma^{(2)}(\omega) = (\omega - \varepsilon_0 - U n_{-\sigma}^{(1)} - \Sigma_\sigma(\omega) + \mathrm{i}\Gamma)^{-1}. \quad (5)$$

From this $G_\sigma^{(2)}(\omega)$, we calculate the second-order electron number $n_\sigma^{(2)} = n[G_\sigma^{(2)}]$. Furthermore, $U n_{-\sigma}^{(1)}$ in the denominator of eq. (5) is replaced by $U n_{-\sigma}^{(2)}$ to find the solutions for $\tilde{\varepsilon}_0$ in a wide range of parameters. We calculate $n_\sigma^{(2)} = n[G_\sigma^{(2)}]$ again and $n_\sigma^{(2)\mathrm{FS}} = n^{\mathrm{FS}}[G_\sigma^{(2)}]$ from the Friedel sum formula,

$$n^{\mathrm{FS}}[G_\sigma] \equiv \frac{1}{2} + \frac{1}{\pi}\tan^{-1}\frac{\mathrm{Re}(G_\sigma(0)^{-1})}{\mathrm{Im}(G_\sigma(0)^{-1})}. \quad (6)$$

Then we determine $\tilde{\varepsilon}_{0\sigma}$ so as to satisfy the relation $n_\sigma^{(2)} = n_\sigma^{(2)\mathrm{FS}}$ (Friedel sum rule).

Figure 1 shows the temperature dependence of the DOS in the dot for U=4, Γ=0.5, ε_0=-2(a) and 0(b). In the case of the symmetry condition ε_0=$-U/2$, side peaks appear at $\omega = \pm 2$ in addition to a Kondo peak at $\omega = 0$ which is supressed by increasing temperature, whereas in the asymmetric case $\varepsilon_0 = 0$, the side peaks disappear and a single peak emerges at a little higher than ω=0.

As we have assumed a single energy level ε_0 for a quantum dot, the current takes the form[11]

$$I = \frac{2e}{\hbar}\sum_\sigma \int d\omega \{f_L(\omega) - f_R(\omega)\}\Gamma' \rho_\sigma(\omega), \quad (7)$$

where $\Gamma' = \Gamma_L\Gamma_R/(\Gamma_L + \Gamma_R)$. For $\rho_\sigma(\omega)$ in eq.(7) we employ the result of Fig. 1 to investigate effects of temperature on dI/dV in a quantum dot. The bias voltage is given by the difference of the chemical potentials between the left and the right leads as $V = \mu_L - \mu_R$. The

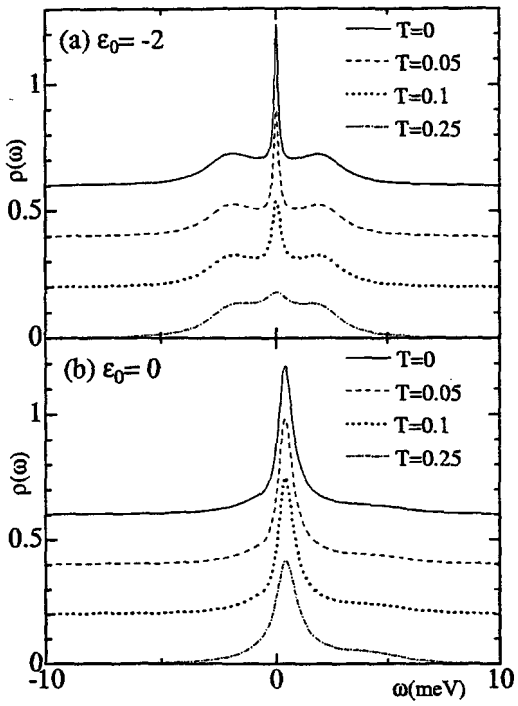

Fig. 1: The calculated DOS at various temperatures are shown.

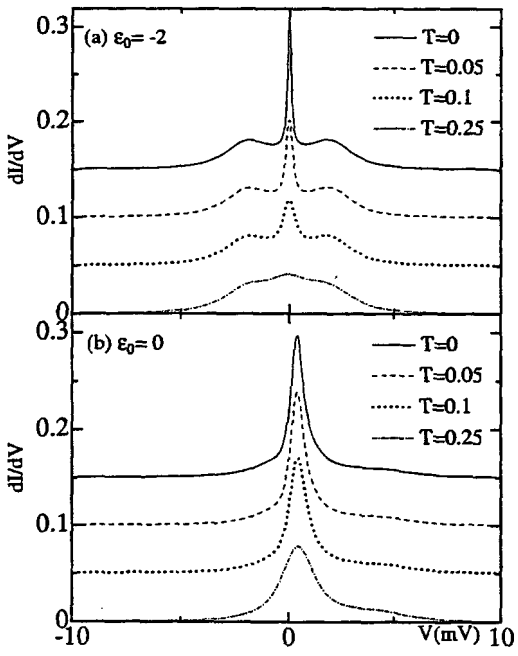

Fig. 2: The calculated dI/dV at various temperatures are shown. The case μ_L=V and μ_R=0.

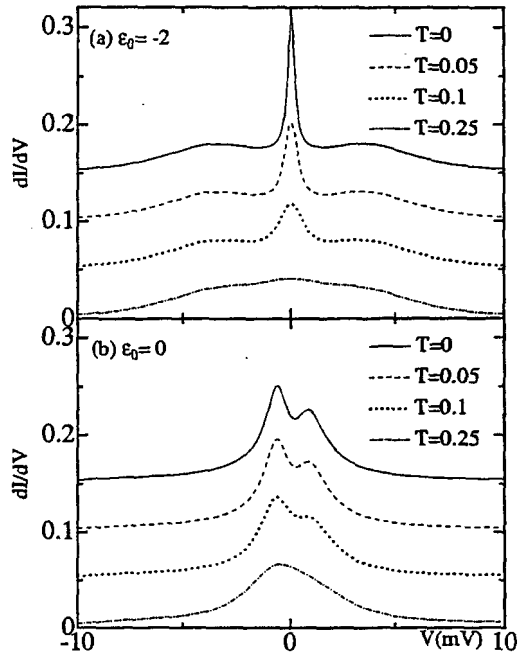

Fig. 3: The calculated dI/dV at various temperatures are shown. The case μ_L=2V/5 and μ_R=−3V/5.

transport properties, however, depend on how it is applied. We investigate the two cases, (1) $\mu_L = V$ and $\mu_R = 0$ and (2) $\mu_L = 2V/5$ and $\mu_R = -3V/5$ when ε_0 is kept unchanged.

Figures 2 (case 1) and 3 (case 2) show the temperature dependence of dI/dV in unit of $2e^2/\hbar$ for $U/\Gamma = 8$. Figure 2 shows that the shape of dI/dV is similar to the result of DOS. Figure 3, however, displays a quite different result, especially in the electron-hole asymmetric case, showing qualitative agreement with the experimental result.[4] These results suggest that we must be careful to extract the DOS from the shape of dI/dV in the electron-hole asymmetry.

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