Gutzwiller dynamic susceptibility: Consequences for the transport properties of transition metals

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We calculate the dynamic susceptibility $\chi(\mathbf{q},\omega)$ of the Hubbard model using the boson representation of Kotliar and Ruckenstein. The energy and momentum dependence of $\chi(\mathbf{q},\omega)$ are the same as in the random-phase approximation, but the Gutzwiller mass enhancement (m^*/m) and spin Landau parameter are included. Combining this with the Kaiser-Doniach expression for the resistivity ρ^{s-d} of a transition metal we obtain a T^2 term in ρ^{s-d} which scales with $(m^*/m)^2$ in accordance with experiment and has the correct magnitude.

INTRODUCTION

Recently we have examined quasiparticle interaction effects in ^{3}He from the standpoint of the "almost localized" description originating in the works of Gutzwiller,[1] Brinkman and Rice[2] on the metal-insulator transition and revived in the context of the liquid state of ^{3}He by Anderson and Brinkman[3] and, more recently, Vollhardt.[4] Starting from the functional integral representation of the Hubbard model introduced by Kotliar and Ruckentein,[5] in which the Gutzwiller approximation appears as a saddle point, we calculated the free energy[6] and two-particle vertex[7] to one-loop order in the fluctuations around the Gutzwiller mean-field result. We obtained the coefficient of the $T^3\ln T$ component in the specific-heat[6] and the superfluid transition temperature[7] as a function of pressure, and obtained satisfactory agreement with experiment for both quantities.

In this paper we examine another response function, namely the dynamic spin susceptibility within the preceding approach. This quantity is of interest not merely in the context of liquid ^{3}He, but also from the point of view of transition-metal magnetism where the Gutzwiller and spin-fluctuation descriptions[8,9] have traditionally been regarded as incompatible. The spin-fluctuation approach to the dynamical spin susceptibility $\chi(\mathbf{q},\omega)$ relies on summing a geometric series of particle-hole diagrams. As the resulting $\chi(\mathbf{q},\omega)$ is peaked at low frequencies and momentum transfer (at least for isotropic systems), the system may be regarded as demonstrating ferromagnetic spin tendencies. Furthermore, these spin fluctuations, which for a transition metal arise predominantly from the d-band, scatter s-p conduction electrons inelastically, giving rise to a resistivity proportional to T^2 at low temperatures.[10] In contrast, the Gutzwiller approach emphasizes the way in which local electron-electron repulsion inhibits electronic motion, increasing the effective mass of the carriers. Incorporating dynamical effects into this picture has hitherto been problematical.

In the following section we calculate $\chi(\mathbf{q},\omega)$ using a novel dynamical mean-field approach. Simply stated, in the presence of a momentum- and frequency-dependent external magnetic field $h(\mathbf{q},\omega)$, the boson fields themselves acquire a $\mathbf{q},\omega$ dependence even at the mean-field level. However, to evaluate the susceptibility only requires a knowledge of the free energy to second order in $h(\mathbf{q},\omega)$, and to this order most of the Bose fields are static and determined by the $h=0$ values. After minimization with respect to the remaining fields, some straightforward algebra is all that is required to obtain $\chi(\mathbf{q},\omega)$. The method is rather more transparent than that based on functional derivatives of the Gaussian free energy (see Ref. 11 for an elaboration of this technique) although both methods lead to the same result.

In Sec. III we examine the resistivity $\rho^{s\text{-}d}$ due to s-d scattering in transition metals using the variational solution of the Boltzmann equation derived by Kaiser and Doniach,[12] with our $\chi(\mathbf{q},\omega)$ as the only input. We calculate the T^2 term in $\rho^{s\text{-}d}$ and discuss the correspondence with experiment.

DYNAMIC SUSCEPTIBILITY

The Hubbard model can be written in the following form:[5]

$$H=\sum_{i,j,\sigma} t_{ij} f_{i\sigma}^{\dagger} f_{j\sigma} z_{i\sigma}^{\dagger} z_{j\sigma} + U\sum_i d_i^{\dagger} d_i - \sum_{i,\sigma} f_{i\sigma}^{\dagger} f_{i\sigma}\mu_{i\sigma} , \tag{1}$$

where $\mu_{i\sigma}=\mu+\sigma h_i$ is the chemical potential modulated by the external inhomogenous and time-dependent field h_i. The spin-σ band electrons at site i are represented by creation operators f_i and have hopping matrix elements t_{ij} while U denotes the on-site Coulomb repulsion. The boson operators e_i, $p_{i\sigma}$, and $d_{i\sigma}$ label the empty, spin-σ and doubly occupied sites, respectively, and are forced to satisfy the following constraints:

$$\sum_\sigma p_{i\sigma}^{\dagger} p_{i\sigma} + e_i^{\dagger} e_i + d_i^{\dagger} d_i = 1 , \tag{2}$$

$$f_{i\sigma}^{\dagger} f_{i\sigma} = p_{i\sigma}^{\dagger} p_{i\sigma} + d_i^{\dagger} d_i , \tag{3}$$

while the hopping factor $z_{i\sigma}$ is defined by

$$z_{i\sigma}=(1-d_i^\dagger d_i-p_{i\sigma}^\dagger p_{i\sigma})^{-1/2}(e_i^\dagger p_{i\sigma}+p_{i-\sigma}^\dagger d_i)(1-e_i^\dagger e_i-p_{i-\sigma}^\dagger p_{i-\sigma})^{-1/2} . \quad (4)$$

The partition function can be written in terms of functional integrals as follows:

$$Z=\prod_{i,\sigma} De_i Dp_{i\sigma} Dd_i d\lambda_i d\lambda_{i\sigma} Df_{i\sigma}^\dagger Df_{i\sigma}\delta(\arg(p_{i\uparrow}+p_{i\downarrow})-\arg(e_i+d_i))\exp\left[-\int_0^\beta d\tau L(\tau)\right] \quad (5)$$

where gauge invariance has been ensured by including into the functional integrations a δ function involving the phases of the Bose fields and the constraints are included via the Lagrangian multipliers $\lambda_i^{(1)}$, $\lambda_{i\sigma}^{(2)}$ and $\mu_{i\sigma}$. The Lagrangian action is written

$$L(\tau)=\sum_i \Bigg\{ e_i^\dagger(\tau)\partial/\partial\tau e_i(\tau)+d_i^\dagger(\tau)(\partial/\partial\tau+U)d_i(\tau)+\sum_\sigma p_{i\sigma}^\dagger(\tau)\partial/\partial\tau p_{i\sigma}(\tau)$$
$$+i\lambda_i^{(1)}\left[\sum_\sigma p_{i\sigma}^\dagger(\tau)p_{i\sigma}(\tau)+e_i^\dagger e_i+d_i^\dagger d_i-1\right]+\sum_{i,\sigma} i\lambda_{i\sigma}^{(2)}[f_{i\sigma}^\dagger(\tau)f_{i\sigma}(\tau)-p_{i\sigma}^\dagger(\tau)p_{i\sigma}(\tau)-d_i^\dagger(\tau)d_i(\tau)]\Bigg\}$$
$$+\sum_{i,j,\sigma} t_{ij}f_{i\sigma}^\dagger(\tau)f_{j\sigma}(\tau)z_{i\sigma}^\dagger(\tau)z_{j_\sigma}(\tau)+\sum_{i,\sigma} f_{i\sigma}^\dagger(\tau)(\partial/\partial\tau-\mu_{i\sigma})f_{i\sigma}(\tau) , \quad (6)$$

which, after the following gauge transformations and changes of variable (see Ref. 13)

$$e_i(\tau)=x_i(\tau)\exp[i\theta_i(\tau)], d_i(\tau)=y_i(\tau)\exp[i\phi_i(\tau)] ,$$
$$p_{i\sigma}(\tau)=q_{i\sigma}(\tau)\exp[i\chi_{i\sigma}(\tau)], f_{i\sigma}(\tau)=f'_{i\sigma}(\tau)\exp[-i\chi_{i\sigma}(\tau)+i\theta_i(\tau)] , \quad (7)$$
$$\alpha_i(\tau)=i\lambda_i^{(1)}(\tau)+i\dot\theta_i(\tau), \beta_{i\sigma}(\tau)=i\lambda_{i\sigma}^{(2)}(\tau)+i\dot\theta_i(\tau)-i\dot\chi_{i\sigma}(\tau) ,$$

becomes, on invoking periodicity of the bose fields, a sum of a fermion part

$$L_f(\tau)=\sum_{i,\sigma} f_{i\sigma}^\dagger(\tau)[\partial/\partial\tau-\mu_{i\sigma}(\tau)+\beta_{i\sigma}(\tau)]f_{i\sigma}(\tau)+\sum_{i,j,\sigma} f_{i\sigma}^\dagger(\tau)f_{j\sigma}(\tau)t_{ij}z_{i\sigma}^\dagger(\tau)z_{j\sigma}(\tau) \quad (8)$$

and a bose part

$$L_b(\tau)=\sum_i \left[\alpha_i(\tau)\left[x_i(\tau)^2+y_i(\tau)^2+\sum_\sigma q_{i\sigma}(\tau)^2-1\right]+Uy_i(\tau)^2-\sum_\sigma \beta_{i\sigma}(\tau)[y_i(\tau)^2+q_{i\sigma}(\tau)^2]\right] . \quad (9)$$

Performing the integration over the Fermi fields and introducing the Fourier transforms on the fields we obtain

$$F_f=-T\sum_\sigma \mathrm{Tr}\ln A_\sigma(k_1,k_2) , \quad (10)$$

where

$$A_\sigma(k_1,k_2)=\left\{(-i\omega-\mu)\delta(k_1,k_2)+T[\beta_\sigma(k_1-k_2)-\sigma h(k_1-k_2)]+T^2\sum_p t_p z_\sigma(k_1-p)z_\sigma(p-k_2)\right\} , \quad (11)$$

and q is shorthand for $(\mathbf{q},\omega)$.

At the mean-field level, and to second order in the magnetic field, it is straightforward to show that only the spin-dependent fields β_σ and q_σ depend on $H(\mathbf{q},\omega)$, all the others being given by their $h=0$ mean-field values. The boson part then becomes

$$F_b=\alpha(x^2+y^2)+\alpha T^2\sum_{\sigma,k_1} q_\sigma(-k_1)q_\sigma(k_1)-y^2\sum_\sigma\beta_\sigma+Uy^2-\alpha-T^3\sum_{k_1k_2}\sum_\sigma q_\sigma(-k_1)\beta_\sigma(k_1-k_2)q_\sigma(k_2) . \quad (12)$$

Minimizing the action with respect to the spin-dependence fields according to the variational principle, we obtain

$$\partial F/\partial\beta_\sigma(-k)=0=\partial F_f/\partial\beta_\sigma(-k)-Ty^2\delta(k)-T^3\sum_{k_2} q_\sigma(k-k_2)q_\sigma(k_2) ,$$
$$\partial F/\partial \mathbf{q}_\sigma(-k)=0=\partial F_f/\partial \mathbf{q}_\sigma(-k)+2\alpha T^2 q_\sigma(k)-2\Upsilon^3\sum_{k_1}\beta_\sigma(k-k_1)q_\sigma(k_1) . \quad (13)$$

Differentiating further with respect to the external field yields

$$\sum_\sigma \sigma[(1/2T^2)\partial^2F_f/\partial h(k)\partial q(-k)+(\alpha-\beta)\partial q_\sigma(k)/\partial h(k)-q\partial\beta_\sigma(k)/\partial h(k)]=0 ,$$
$$\sum_\sigma \sigma[\partial^2F_f/\partial h(k)\partial\beta(-k)-T^2\partial n_\sigma(k)/\partial h(k)]=0 . \quad (14)$$

Inserting the expression for F_f together with the relevant mean-field values yields, in the half-filled limit,[5] to

$$\sum_\sigma \sigma \partial\beta_\sigma(k)/\partial h(k) = -U[(1-I)(2+I)/(1+I)][\partial n_\uparrow(k)/\partial h(k) - \partial n_\downarrow(k)/\partial h(k)] ,$$
$$\sum_\sigma \sigma \partial n_\sigma(k)/\partial h(k) = T\sum_{k_1} H(k_1)H(k+k_1)\left[\sum_\sigma \sigma\partial\beta_\sigma(k)/\partial h(k) - 2\right] , \quad (15)$$

in which $I=U/U_c$, and the mean-field propagator is given by

$$H(k)=[-i\omega-\mu_\sigma+\beta_\sigma+z^2 t_k]^{-1} , \quad (16)$$

where t_k denoted the band energy, the mean-field mass enhancement is given by

$$z^2=[1-(U/U_c)^2]^{-1} ,$$

and

$$U_c=16T\sum_{k<k_F,\omega} t_k H(k)$$

denotes a critical value of the coupling at which the effective mass becomes infinite. The field-dependent occupation number is defined by

$$n_\sigma^\sigma(k)=T\sum_{k_1} q_\sigma(k-k_1)q_\sigma(k_1)+y^2/T . \quad (17)$$

Introducing the mean-field Lindhard susceptibility $\chi_0(\mathbf{q},\omega)$,

$$\chi_\sigma(q)=-T\sum_{k_1} H(q+k_1)H(k_1) , \quad (18)$$

we obtain for the full spin susceptibility $\chi(\mathbf{q},\omega)$ defined by

$$\chi(\mathbf{q},\omega)=\sum_\sigma \sigma\partial n_\sigma(\mathbf{q},\omega)/\partial h(\mathbf{q},\omega) , \quad (19)$$

the following form after some manipulation:

$$\chi(\mathbf{q},\omega)=\chi_0(\mathbf{q},\omega)/\{1+[f_0^a/N^*(0)]\chi_0(\mathbf{q},\omega)\} , \quad (20)$$

where $N^*(0)$ is the renormalized density of states at the Fermi level and the Gutwiller spin Landau parameter is given by

$$f_0^a=-pI(2+I)/(1+I)^2 . \quad (21)$$

Our expression for $\chi(\mathbf{q},\omega)$ has the familiar random-phase approximation (RPA) form but with the mean-field spin Landau parameter f_0^a (Ref. 15) entering the denominator. Consequently, since the spin Landau parameter tends to a value of $-3p/4$ [where $p=N(0)\sum_{k<k_F} t_k$ is the only parameter that depends on the details of the bandstructure], as $U\rightarrow U_c$ the system experiences spin fluctuations that are quite long lived but not critical unless $p\geq\frac{4}{3}$. The spectral density $\mathrm{Im}\chi(\mathbf{q},\omega)$ has the familiar peak at small $\mathbf{q}$ and ω, but with a Fermi velocity renormalized by the Gutzwiller factor $(1-U/U_c)^2$. We note that for small U the spin Landau parameter approaches $UN(0)$ and the standard RPA result is recovered.

RESISTIVITY OF TRANSITION METALS

To obtain the resistivity contribution arising from exchange scattering of s-p electrons off the d electrons, we substitute $\chi(\mathbf{q},\omega)$ obtained earlier into the Kaiser-Doniach expression,[12] modified by Jullien *et al.*[14] to allow for different s-p and d-electron Fermi momenta (k_{F_c} and k_{F_d} respectively)

$$\rho=(\rho_0/T)\int_0^\infty d\omega\,\omega A(\omega)/\{[\exp(\beta\omega)-1]\times[1-\exp(-\beta\omega)]\} , \quad (22)$$

where $A(\omega)$ denotes the imaginary part of the $\mathbf{q}$-averaged, d-electron spin susceptibility

$$A(\omega)=(2/k_{F_c}{}^4)\int_0^{2k_{F_c}} dq\, q^3\mathrm{Im}\chi(\mathbf{q},\omega) , \quad (23)$$

and the constant ρ_0 may be written as

$$\rho_0=[JN_e(0)/4]^2(\nu_e/n_e)[m_e/(n_e e^2\tau_{F_c})] , \quad (24)$$

where $N_e(0)$ is the s-p density of states and J is the s-p-d exchange constant. The number of s-p electrons (atoms) per unit volume is denoted by $n_e(\nu_e)$, m_e is the conduction (s-p) electron mass, and the inverse lifetime for conduction electrons $1/\tau_{F_c}$ is equal to their Fermi energy. We can simplify matters by looking first at the high-temperature behavior of the resistivity. Although the mean-field boson theory treatment given here is unlikely to be reliable for all temperatures, the theory has a simple high-temperature limit in which the effects of correlation vanish completely and the susceptibility is given by the Curie law

$$\chi(\mathbf{q},\omega=0)=1/(2T) .$$

Following Jullien *et al.*,[14] the Kramers-Kronig relation may be invoked leading to the result that the resistivity saturates at a value $\rho_\infty=2\pi\rho_0$. In fact ρ_∞ will prove a more convenient resistivity scale for our purposes.

The T^2 term in $\rho^{s\text{-}d}$ is obtained by expanding $A(\omega)$ to leading order in ω so that

$$\rho=\rho_\infty(\pi^2/2)I_1[\xi,f_a](m^*/m)^2(T/\epsilon_{F_d})^2 , \quad (25)$$

where ϵ_{F_d} denotes the bare d-band Fermi energy and

$$I_1[\xi,f_a]=(1/\xi^4)\int_0^{\min(\xi,1)} dx\, x^2/[1+f_a\chi(2k_{F_d}x,\omega)/N^*(0)]^2 \quad (26)$$

involves the ratio $\xi(=k_{F_c}/k_{F_d})$ and the spin Landau parameter. For a parabolic band with $\xi=1$, this factor ranges from 2.4 for $f_0^a=0.7$ to 5.2 for $f_0^a=1$. The essential point to note is that the T^2 term scales with the square of the effective mass enhancement as I_1 depends only weakly on U/U_c compared with m^*/m. The scaling of A with $(m^*/m)^2$ was pointed out long ago by Rice[15] who invoked Baber scattering to explain this. In the context of the heavy-fermion (HF) systems several authors have obtained this scaling law.[16] Both these approaches and the one given here have in common the scattering of fermions by bosons representing the long-wavelength modes of the system. However in the HF theories the scattered electrons play a part in the heavy band formation while in the present problem the dominant resistivity arises from light electrons weakly coupled to the strongly correlated band. Consequently, the ratio ρ/T^2 to m^*/m is expected to differ between the two systems. We can compare with experiment more readily if we introduce the specific-heat coefficient

$$\gamma=2\pi^2N^*(0)/3=\pi^2/(2\epsilon_{F_d}{}^*)\ ,$$

where $\epsilon_{F_d}{}^*=\epsilon_{F_d}m/m^*$ is the effective d-electron bandwidth. Choosing units such that $k_B=1$, we find that the ratio of $A\ (=\rho/T^2)$ to γ^2 is given by

$$A/\gamma^2=\rho_\infty(2I_1[\xi,f_0]/\pi^2)\ . \qquad (27)$$

We may compare this with the ratio obtained in heavy fermion theories, e.g., that of Coleman,[16] where for a spin-$\frac{1}{2}$ system $A/\gamma^2=9\rho_U/4$, with ρ_U denoting the unitary scattering resistance. Setting ρ_U equal to 350 $\mu\Omega$ cm yields the observed value for HF systems

$$A/\gamma^2=10^{-5}\ \mu\Omega\,\text{cm}\,\text{mJ}^{-2}\,\text{mol}^2\,\text{K}^4\ .$$ [17]

For our problem it is ρ_∞ rather than ρ_U which determines the size of A/γ^2 and from experiments on Pd (see Ref. 14 for references) we take ρ_∞ to be 50 $\mu\Omega$ cm. Setting $f_0^a=-0.9$ and taking $\xi=1$ we obtain (assuming a parabolic band)

$$A/\gamma^2=0.5\times10^{-6}\ \mu\Omega\,\text{cm}\,\text{mJ}^{-2}\,\text{mol}^2\,\text{K}^4$$

in reasonable agreement with the experimental value for transition metals given by Rice.[15] More recent measurements of A for Pd (Ref. 18) are still in accord with this value.

In summary, we have calculated the Gutzwiller dynamic susceptibility of the Hubbard model starting from the boson representation of Kotliar and Ruckenstein.[5] For a parabolic band the response is peaked at low **q** and ω but with an energy scale renormalized according to the Gutwiller approximation. Taking $\chi(\mathbf{q},\omega)$ as input to a standard formula describing s-d scattering in transition metals, we obtain a T^2 component in the resistivity which scales with the square of the mass enhancement in agreement with long-standing experiments. The ratio A/γ^2 is smaller than in HF systems by an order of magnitude, reflecting the fact that light s-p electrons dominate the resistivity.

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