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Magnetic susceptibility of the double-exchange model

R. S. Fishman
Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6032

M. Jarrell
Department of Physics, University of Cincinnati, Cincinnati, Ohio 45221

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A unified treatment is provided for the local-moment, electronic, and cross terms in the magnetic susceptibility χ of the double-exchange model within the dynamical mean-field theory. All contributions diverge at the same critical temperature T_C, which may be calculated analytically for large $J_H S$ and a semi-circular density-of-states. Close to T_C, the Curie constant deviates significantly from the result expected for a local-moment system.

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Over the past decade, itinerant theories of magnetism like the double-exchange (DE) models have become increasingly important to address the complex behavior of oxide materials. One of the most powerful tools for studying itinerant models is dynamical mean-field theory (DMFT),[1] which may be considered as the generalization of conventional mean-field theory for itinerant rather than local-moment systems. Yet despite the great success of DMFT in explaining the physics of the DE model,[2–4], a complete theory for the magnetic susceptibility is lacking. While theories of the local-moment[5] and electronic[6] contributions to the susceptibility have recently been used to evaluate the Curie temperature T_C, no such theory exists for the cross terms and the total susceptibility χ has never been explicitly evaluated. In this paper, we place all of the contributions to the susceptibility on an equal footing and calculate χ within a matrix formulation. For a highly-correlated system with large Hund's coupling, the Curie constant deviates from the result expected for a local-moment system.

The Hamiltonian of the DE model is

$$H=-t\sum_{\langle i,j\rangle}(c^{\dagger}_{i\alpha}c_{j\alpha}+c^{\dagger}_{j\alpha}c_{i\alpha})-2J_H\sum_i \mathbf{s}_i\cdot\mathbf{S}_i, \qquad (1)$$

where $c^{\dagger}_{i\alpha}$ and $c_{i\alpha}$ are the creation and destruction operators for an electron with spin α at site i, $\mathbf{s}_i=(1/2)c^{\dagger}_{i\alpha}\vec{\sigma}_{\alpha\beta}c_{i\beta}$ is the electronic spin, $\mathbf{S}_i=S\hat{\mathbf{m}}_i$ is the spin of the local moment, and J_H is the Hund's coupling between the local and electronic spins. As is customary, the local moments are treated classically. However, we shall delay taking the limit of large $J_H S$ or specializing to a particular case for the noninteracting density-of-states (DOS) $\rho_0(\epsilon)$ so that our general results apply to both hypercubic (Gaussian DOS) and Bethe (semicircular DOS) lattices.

Within the DE model, the local moments interact through the hopping of electrons between neighboring sites. Due to the close connection between electron conduction and magnetic order, the double-exchange model qualitatively explains the metal-insulator transition observed in manganites like $La_{1-x}Sr_xMnO_3$ when $x\approx 0.3$.[3] When $J_H S$ exceeds a critical value proportional to the band width W,[7] the interacting DOS contains two bands: a lower band where the spins of the electrons and local-moments at each site are parallel and an upper band where they are antiparallel. So the system is an insulator when the band filling $p=1-x$ (number of electrons per site) equals 1 and the lower band is completely full.

In imaginary time, the full magnetic susceptibility of the DE model is given by

$$\chi(\mathbf{q},i\omega_m)_{\alpha\beta}=\int_0^{\beta}d\tau e^{i\omega_m\tau}\sum_i e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_1)}\times\langle T_\tau S^{\alpha}_{\mathrm{tot},i}(\tau)S^{\beta}_{\mathrm{tot},1}(0)\rangle, \qquad (2)$$

where $\omega_m=2m\pi T$, T_τ is the time-ordering operator, and $\mathbf{S}_{\mathrm{tot},i}=\mathbf{S}_i+\mathbf{s}_i$ is the total spin at site i. The isotropy of the susceptibility $\chi(\mathbf{q},i\omega_m)_{\alpha\beta}=\chi(\mathbf{q},i\omega_m)\delta_{\alpha\beta}$ above T_C is used to set $\alpha=\beta=z$. It is apparent that $\chi(\mathbf{q},i\omega_m)$ contains three sets of terms: the local-moment susceptibility, the electronic susceptibility, and the cross terms. Of course, our goal is to evaluate the elastic susceptibility $\chi=\lim_{q\to 0,\omega\to 0}\chi(\mathbf{q},i\omega_m\to\omega+i\varepsilon)$.

Since the electronic susceptibility contains two Fermion sums, the cross terms one sum, and the local-moment term no sum, their contributions to the full susceptibility may be written as

$$\chi^{ss}(\mathbf{q},i\omega_m)=\int_0^{\beta}d\tau e^{i\omega_m\tau}\sum_i e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_1)}\langle T_\tau s^z_i(\tau)s^z_1(0)\rangle=\sum_{l,n}\chi^{ss}_{ln}(\mathbf{q},i\omega_m), \qquad (3)$$

$$\chi^{Ss}(\mathbf{q},i\omega_m)=\int_0^{\beta}d\tau e^{i\omega_m\tau}\sum_i e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_1)}\langle T_\tau S^z_i(\tau)s^z_1(0)\rangle=\sum_l\chi^{Ss}_l(\mathbf{q},i\omega_m), \qquad (4)$$

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$$\chi^{sS}(\mathbf{q},i\omega_m)=\int_0^\beta d\tau e^{i\omega_m\tau}\sum_i e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_1)}\langle T_\tau s_i^z(\tau)S_1^z(0)\rangle$$

$$=\sum_l \chi_l^{sS}(\mathbf{q},i\omega_m), \tag{5}$$

$$\chi^{SS}(\mathbf{q},i\omega_m)=\int_0^\beta d\tau e^{i\omega_m\tau}\sum_i e^{-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_1)}\langle T_\tau S_i^z(\tau)S_1^z(0)\rangle, \tag{6}$$

where, for example, $\chi_{ln}^{ss}(\mathbf{q},i\omega_m)$ is a matrix with indices l and n denoting the internal Fermi Matsubara frequencies $\nu_{l,n}=(2n_{l,n}+1)\pi T$. Because the local moments are classical quantities which commute with H, $S_i^z(\tau)=S_i^z$ and $\chi^{Ss}(\mathbf{q},i\omega_m)=\chi^{sS}(\mathbf{q},i\omega_m)$. The full susceptibility $\chi(\mathbf{q},i\omega_m)$ may be evaluated by summing over all matrix elements of

$$\underline{\chi}(\mathbf{q},i\omega_m)=\begin{pmatrix}\chi^{SS}(\mathbf{q},i\omega_m) & \underline{\chi}^{Ss}(\mathbf{q},i\omega_m)\\ \underline{\chi}^{sS}(\mathbf{q},i\omega_m) & \underline{\chi}^{ss}(\mathbf{q},i\omega_m)\end{pmatrix}. \tag{7}$$

If the Fermi frequencies are restricted to values $|\nu_{l,n}|\leq(2n_\chi-1)\pi T$, then $\underline{\chi}^{ss}$ is a $2n_\chi\times 2n_\chi$ matrix, $\underline{\chi}^{Ss}$ and $\underline{\chi}^{sS}$ are $2n_\chi$-dimensional row and column vectors, respectively, and the full susceptibility matrix $\underline{\chi}$ has dimensions $(2n_\chi+1)\times(2n_\chi+1)$. We shall take the limit $n_\chi\to\infty$ before presenting our final results.

For a fixed ω_m, $\underline{\chi}(\mathbf{q},i\omega_m)$ obeys the Bethe-Salpeter equation[8]

$$\underline{\chi}(\mathbf{q},i\omega_m)=\underline{\chi}^{(0)}(\mathbf{q},i\omega_m)+\underline{\chi}^{(0)}(\mathbf{q},i\omega_m)\underline{\Gamma}(i\omega_m)\underline{\chi}(\mathbf{q},i\omega_m), \tag{8}$$

where $\Gamma(i\omega_m)$ is the vertex function and $\chi^{(0)}(\mathbf{q},i\omega_m)$ is the bare susceptibility. Within DMFT, momentum conservation at the internal vertices of irreducible graphs is disregarded so that internal Green's functions are replaced by their local values. Consequently, $\Gamma(i\omega_m)$ is independent of momenta and may be evaluated from an identical Bethe-Salpeter equation where $\chi(\mathbf{q},i\omega_m)$ and $\chi^{(0)}(\mathbf{q},i\omega_m)$ are replaced by *local* susceptibilities at site 1, obtained from the $\mathbf{q}$-dependent susceptibilities by summing over all $\mathbf{q}$. In other words, $\Gamma(i\omega_m)=\chi^{(0)-1}(\mathbf{q},i\omega_m)-\chi^{-1}(\mathbf{q},i\omega_m)=\chi^{\mathrm{loc}(0)-1}(i\omega_m)-\chi^{\mathrm{loc}-1}(i\omega_m)$. Like the full susceptibility matrix $\underline{\chi}$, $\underline{\Gamma}$ also has dimensions $(2n_\chi+1)\times(2n_\chi+1)$. Because we are interested only in the elastic susceptibility, we shall henceforth take $\omega_m=0$ and $\mathbf{q}=0$.

To calculate the bare terms in the susceptibility, the electron Green's function and local moments are explicitly decoupled. Since $\langle\mathbf{S}_i\rangle=0$, it follows that $\chi^{sS(0)}=\chi^{Ss(0)}=0$. Using the relation $\langle(S_i^z)^2\rangle=S^2/3$, we easily obtain the local susceptibility matrices

$$\underline{\chi}^{\mathrm{loc}(0)}=\begin{pmatrix}S^2/3T & 0\\ 0 & \underline{\chi}^{ss\mathrm{loc}(0)}\end{pmatrix}, \tag{9}$$

$$\underline{\chi}^{\mathrm{loc}}=\begin{pmatrix}S^2/3T & S\underline{g}/3\\ S\underline{g}^{\mathrm{T}}/3 & \underline{\chi}^{ss\mathrm{loc}}\end{pmatrix}, \tag{10}$$

where $\chi^{Ss\mathrm{loc}}=(S/3)\Sigma_l g_l$ or

$$g_l=-\frac{3}{2S}\int_0^\beta d\tau e^{i\nu_l\tau}\langle T_\tau c_{1\beta}(\tau)c_{1\alpha}^\dagger(0)S_1^z\rangle\sigma_{\alpha\beta}^z. \tag{11}$$

With the additional definitions $\kappa=1-T\underline{g}^{\mathrm{T}}\underline{\chi}^{ss\mathrm{loc}-1}\underline{g}/3$ and $\delta_l=T\Sigma_n g_n\chi_{nl}^{ss\mathrm{loc}-1}$, it is straightforward to show that the vertex function has matrix elements $\Gamma^{SS}=(3T/S^2)(1-1/\kappa)$, $\Gamma_l^{Ss}=\Gamma_l^{sS}=\delta_l/S\kappa$, and $\Gamma_{ln}^{ss}=\chi_{ln}^{ss\mathrm{loc}(0)-1}-\chi_{ln}^{ss\mathrm{loc}-1}-\delta_l\delta_n/3T\kappa$.

Fermion lines in the bare, $\mathbf{q}=0$ susceptibility do not connect local moments on different sites so that

$$\underline{\chi}^{(0)}(\mathbf{q}=0)=\begin{pmatrix}S^2/3T & 0\\ 0 & \underline{\chi}^{ss(0)}(\mathbf{q}=0)\end{pmatrix}. \tag{12}$$

Consequently, the inverse of the full, $\mathbf{q}=0$ susceptibility matrix is

$$\underline{\chi}^{-1}(\mathbf{q}=0)=\begin{pmatrix}3T/\kappa S^2 & -\underline{\delta}/S\kappa\\ -\underline{\delta}^{\mathrm{T}}/S\kappa & \underline{\chi}^{\mathrm{el}-1}(\mathbf{q}=0)+\underline{\delta}\,\underline{\delta}/3T\kappa\end{pmatrix}, \tag{13}$$

where

$$\underline{\chi}^{\mathrm{el}-1}(\mathbf{q}=0)=\underline{\chi}^{ss(0)-1}(\mathbf{q}=0)-\underline{\chi}^{ss\mathrm{loc}(0)-1}+\underline{\chi}^{ss\mathrm{loc}-1} \tag{14}$$

is the inverse of the $\mathbf{q}=0$ electronic susceptibility matrix evaluated from its *own* Bethe-Salpeter equation.[6]

The condition that $\chi\to\infty$ at T_C is equivalent to the condition $\mathrm{Det}(\underline{\chi}^{-1}(\mathbf{q}=0))\to 0$. Using Eq. (13) for $\underline{\chi}^{-1}(\mathbf{q}=0)$, we derive

$$\mathrm{Det}(\underline{\chi}^{-1}(\mathbf{q}=0))=\frac{3T}{\kappa S^2}\mathrm{Det}(\underline{\chi}^{\mathrm{el}-1}(\mathbf{q}=0)). \tag{15}$$

Since each matrix element of $\underline{\chi}(\mathbf{q}=0)$ is inversely proportional to $\mathrm{Det}(\underline{\chi}^{-1}(\mathbf{q}=0))$, all contributions to the susceptibility diverge at the same critical temperature. It also follows that the electronic susceptibility drives the divergences of the local-moment and cross terms in the full susceptibility!

To make this analysis more concrete, we now specialize to a Bethe lattice with noninteracting DOS given by the semicircular form $\rho_0(\epsilon)=(8/\pi W^2)\sqrt{W^2/4-\epsilon^2}$, where $W=4t\sqrt{z}$ is the full band width. We also take the limits $J_{\mathrm{H}}S\gg W$ and $J_{\mathrm{H}}S\gg T$ so that the upper and lower bands in the interacting DOS are well separated and the mobile carriers are restricted to one of those two bands. When $|\nu_l|\ll J_{\mathrm{H}}S$, it is easy to show that $g_l\to s_p/f_l$, where $s_p=\mathrm{sgn}(p-1)$ depends on whether p is greater or less than 1 and $f_l=z_l+\sqrt{z_l^2-W^2/8}$ with $z_l=i\nu_l+\delta\mu$ and $\delta\mu=\mu-s_pJ_{\mathrm{H}}S$. Using the local Green's functions first derived by Furukawa[2,3] for a fixed direction $\hat{\mathbf{m}}_i$ of the Mn moment, the local electronic susceptibility involves the average of two local Green's functions with respect to $\hat{\mathbf{m}}_i$. It can then be shown that[6]

$$\chi_{ln}^{ss\mathrm{loc}-1}=\frac{3}{T}\left\{-\delta_{ln}f_l^2+\frac{f_lf_n}{2n_\chi-1}\right\}. \tag{16}$$

Hence, $\kappa=-1/(2n_\chi-1)$, $\delta_l=3f_l s_p/(2n_\chi-1)$, and the total susceptibility is given by

$$\chi=\frac{1}{3T}\frac{S_{eff}(T)^2}{1-F_1(T)}+\frac{8T}{W^2}F_1(T), \tag{17}$$

$$S_{eff}(T)=S+\frac{1}{2}-\frac{1}{2}|p-1|+s_pF_2(T). \tag{18}$$

Here, $F_1(T)$ and $F_2(T)$ are defined by the Fermion sums

$$F_1(T)=\sum_l \frac{R_l^2}{R_l^2-3W^2/32}, \tag{19}$$

$$F_2(T)=T\sum_l \frac{R_l^2}{(R_l^2-3W^2/32)f_l}, \tag{20}$$

where $R_l=f_l/2-z_l$ and the restriction on the Fermi frequencies ν_l has been lifted. Since $\chi^{SS}\propto S^2$, $\chi^{sS}\propto S$, and $\chi^{ss}\propto S^0$, it is simple to separate the contributions of the local moments, cross terms, and electrons to the full susceptibility.

Each of these contributions diverges as $F_1(T)\to 1$, so T_C is solved from the condition

$$\sum_l \frac{R_l^2}{R_l^2-3W^2/32}=1. \tag{21}$$

Also in the limit of large $J_H S$, the band filling is

$$p=1+\frac{s_p}{2}+2T\sum_l \mathrm{Re}\left\{\frac{1}{f_l}\right\}. \tag{22}$$

These results correct an error by Furukawa[9] but agree in the limit $T_C\ll W$ with Auslender and Kogan.[5] The resulting T_C is symmetric about $p=0.5$ and $p=1$, and peaks at $p=0.5$ with $T_C\approx 0.022W$, and vanishes at $p=0$ and $p=1$. When $p=1$, T_C vanishes because electrons are unable to hop to neighboring sites without incurring an enormous cost in Hund's energy. The final term in Eq. (17) for χ is part of the electronic susceptibility that does not diverge at T_C. For a given temperature, $8TF_1(T)/W^2\approx 8T_C/W^2$ peaks at $p=0.5$ or 1.5, when the number of mobile carriers is largest. Hence, this term is nothing but the Pauli susceptibility of the electrons in the limit of large $J_H S$.

Based on the mean-field (MF) theory[10] of local moments with spin S_0, the expected Curie-Weiss (CW) result for the magnetic susceptibility is $\chi=C/(T-T_C)$ with Curie constant $C=S_0^2/3$. Our result for χ in Eq. (17) deviates from the MF prediction in two ways: both the effective spin $S_{eff}(T)$ and the denominator $T[1-F_1(T)]$ provide nontrivial corrections to the Curie constant. We shall first discuss these corrections for general S.

As implied by Eq. (18), the effective spin of the coupled electronic and local-moment system equals $S_{eff}(T)=S_0+s_pF_2(T)$ where $S_0=S+1/2-|p-1|/2$ is the[11] expected result for strong Hund's coupling: $S_0=S$ when $p=0$ and 2 (no electrons per site or two electrons with opposite spins on each site) and $S_0=S+1/2$ for $p=1$ or one electron per site with spin parallel to that of the local moment. The difference S_0-S is plotted versus p in the dashed lines of the inset to Fig. 1. Evidently, $s_pF_2(T)$ provides a correction to the effective spin due to strong electronic correlations. Evaluated at T_C, $F_2(T_C)$ vanishes when $p=0$ or 1 and also for $p=1/2$. It is negative for $0\leqslant p\leqslant 1/2$ or $\delta\mu<0$ and positive for $1/2\leqslant p\leqslant 1$ or $\delta\mu>0$, reaching maximum or minimum values of ± 0.03 for $p\approx 0.85$ or 0.15. The effective spin $S_{eff}(T_C)$ at T_C, given by the solid curve in the inset to Fig. 1, is slightly smaller than S_0 for $1/2<p<1$ and slightly larger for $0<p<1/2$. As $T/W\to\infty$, $F_2(T)\to 0$ and $S_{eff}(T)\to S_0$ so that the expected value for the effective spin is restored. Both S_0 and $S_{eff}(T)$ are symmetric about $p=1$.

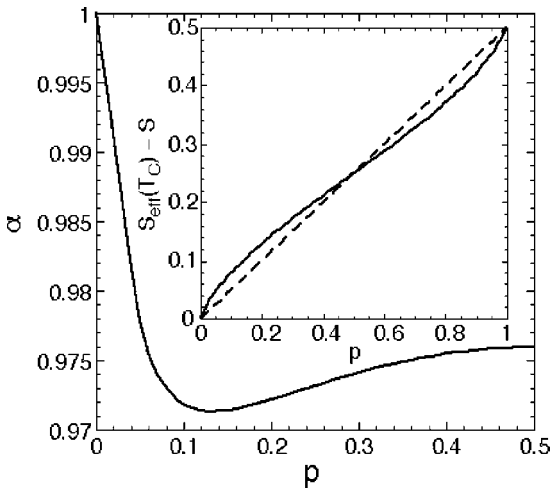

FIG. 1. The renormalization factor α versus band filling p between 0 and 1/2. Symmetric results are obtained for $1/2\leqslant p\leqslant 1$. Inset is the effective spin $S_{eff}(T)$ evaluated at T_C (solid) or at high temperatures $T\gg W$ (dashed) versus band filling p. Both sets of results are for a semicircular DOS with large $J_H S$.

The susceptibility deviates from the MF result in another respect. Linearized near T_C, the inverse local-moment susceptibility may be written in the CW form: $1/\chi^{SS}=3T(1-F_1(T))/S^2\approx 3(T-T_C)/\alpha S^2$ with Curie constant $C=\alpha S^2/3$. Of course, the MF theory of local-moment systems gives $\alpha=1$. But in the DE model, the interactions between the local moments are mediated by highly-correlated electrons and this identity fails. We have evaluated α by linearizing $TF_1(T)$ about T_C. As shown in Fig. 1 for $0\leqslant p\leqslant 1/2$, $\alpha\leqslant 1$ and approaches 1 only as $p\to 0$. The renormalization factor α deviates from unity by as much as 2.8% when $p\approx 0.13$. So even in the limit of large S, χ differs appreciably from the MF prediction for a local-moment system. However, the expected value for the Curie constant is recovered as $T/W\to\infty$ and $\chi^{SS}\to S^2/3T$, the appropriate limit for noninteracting spins. Also note that in the opposite limit $T\ll W$ assumed by Auslender and Kogan,[5] the temperature dependence of $TF_1(T)$ disappears and $\alpha\to 1$.

These predictions are probably best applied to the manganite $La_{1-x}Sr_xMnO_3$ with $x\approx 1/3$, which has a high Curie temperature of 350 K and is believed to be well described by the simple DE model.[3] In the temperature range $T_C\leqslant T\ll W$, our result for χ^{-1} may be linearized as $1/\chi\approx(T$

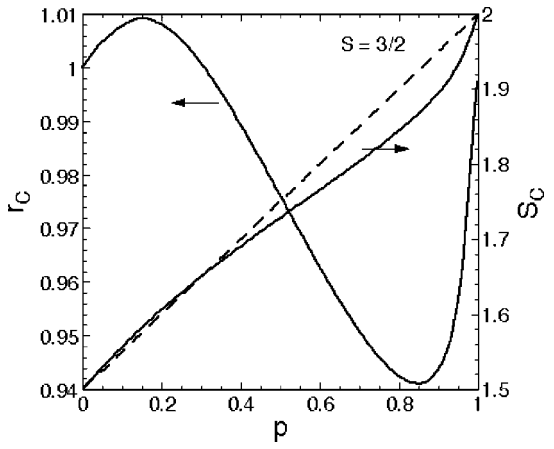

FIG. 2. The renormalization factor r_C (left axis) and Curie spin S_C (right axis), which parametrize the linearized inverse susceptibility, for a system with $S=3/2$. Also plotted in the dashed line is S_0 (right axis).

$-T_C)/C$, where $C=r_C S_0^2/3$ and $r_C=\alpha[S_{eff}(T_C)/S_0]^2$ renormalizes the Curie constant. Specializing to the case $S=3/2$, r_C is plotted versus p in Fig. 2. After an initial increase for small p, r_C falls below one with a minimum value of about 0.941 when $p=0.85$. Rewriting $C=S_C^2/3$ defines the Curie spin $S_C=\sqrt{\alpha}S_{eff}(T_C)=\sqrt{r_C}S_0$, which is also plotted in Fig. 2 and is a monotonically increasing function of the electronic filling p. For a manganite with $x=1/3$ or $p=2/3$, the Curie constant and Curie spin are, respectively, about 4.5% and 2.4% smaller than predicted by MF theory for a local-moment system with spin $S_0=1.833$. Keep in mind, however, that our analysis neglects the quantum corrections associated with the finite size of S.

Unfortunately, the most extensive susceptibility measurements are for the manganite $La_{1-x}Ca_xMnO_3$ with $x=1/3$, which has a lower Curie temperature of 250 K and is not as well-described by the simple DE model. The measured susceptibility[12] between T_C and $2.3T_C$ is much more complex than predicted, with a bulge just above T_C and a ''high-temperature'' limit (above about $1.8T_C$) close to $3(T-\Theta)/S_0^2$, where $\Theta\approx 1.25T_C$. Our model cannot explain the behavior in the critical regime very close to T_C because DMFT does not support long-range correlations. Moreover, magnetic polarons have a strong effect on the susceptibility up to about $1.8T_C$. Nonetheless, it would be interesting to quantitatively compare the predictions of our model with the measured slope of $1/\chi$ in the linear regime between $1.8T_C$ and $2.3T_C$, which still falls within the low-temperature $T \ll W$ range of our model.

To summarize, we have developed a unified treatment of all terms in the magnetic susceptibility of the DE model. As expected, the local-moment, electronic, and cross terms all diverge at the same critical temperature. For the case of a Bethe lattice with large $J_H S$, we have evaluated the susceptibility analytically and shown that the Curie constant is altered by electronic correlations close to T_C. Our work has also demonstrated that DMFT can be used to evaluate the contributions to the magnetic susceptibility of itinerant systems in a systematic and compact fashion.

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[10] See, for example, K. Yosida *Theory of Magnetism* (Springer, Berlin, 1991).
[11] The result for S_{eff} uses the condition

$$T\sum_l g_l=\frac{1}{4}-Ts_p\sum_l \mathrm{Re}\left\{\frac{1}{f_l}\right\}=\frac{1}{2}-\frac{1}{2}|p-1|=S_0-S, \quad (23)$$

which vanishes at $p=0$ and 2.
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