Monte Carlo Method for Magnetic Impurities in Metals

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We discuss a Monte Carlo algorithm to study properties of dilute magnetic alloys; the method can treat a small number of magnetic impurities interacting with the conduction electrons in a metal. Results for the susceptibility of a single Anderson impurity in the symmetric case show the expected universal behavior at low temperatures. Some results for two Anderson impurities are also discussed.

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This paper discusses a Monte Carlo method to study properties of a small number of magnetic impurities interacting with the conduction electrons in a metal. As usual, we assume that electron-electron interactions occur only at the impurity sites. Our algorithm is related to the method introduced by Blankenbecler, Scalapino, and Sugar[1] (BSS) to study fermion systems on a lattice, but has a number of advantages for the study of magnetic impurity systems: It can deal with an infinite sea of conduction electrons, and it does not suffer from instabilities at low temperatures. Although the BSS algorithm can also be used for magnetic impurities,[2] it is not possible to reach with it the interesting low-temperature regime as a result of these instabilities.[2]

Consider for definiteness the single-impurity Anderson model:

$$H=\sum_{k\sigma}\epsilon_k c^\dagger_{k\sigma}c_{d\sigma}+\sum_{k,\sigma}V_k(c^\dagger_{k\sigma}d_\sigma+\text{H.c.})+\epsilon_d\sum_\sigma n_{d\sigma}+Un_{d\uparrow}n_{d\downarrow}\equiv H_0+H_1. \tag{1}$$

In a discrete path-integral formulation the partition function is

$$Z=\mathrm{Tr}e^{-\beta H}=\mathrm{Tr}\prod_{l=1}^{L}e^{-\Delta\tau H}\cong\mathrm{Tr}\prod_{l=1}^{L}e^{-\Delta\tau H_0}e^{-\Delta\tau H_1} \tag{2}$$

with $\beta=L\Delta\tau$. We decouple the interaction part of the Hamiltonian by introducing auxiliary Ising variables[3]:

$$\exp(-\Delta\tau H_1)=\exp\{-\Delta\tau U[n_{d\uparrow}n_{d\downarrow}-\tfrac{1}{2}(n_{d\uparrow}+n_{d\downarrow})]\}=\tfrac{1}{2}\mathrm{Tr}_\sigma\exp[\lambda\sigma(n_{d\uparrow}-n_{d\downarrow})] \tag{3}$$

with $\cosh\lambda=\exp(\Delta\tau U/2)$, and take the trace over fermion degrees of freedom. The result can be written as

$$Z=\mathrm{Tr}_\sigma\prod_{\mu=\pm1}\det{}_{N,L}O_\mu(\{\sigma_l\}), \tag{4}$$

where O_μ is an $NL\times NL$ matrix, with N the number of spatial sites (or k vectors) for the conduction electrons plus 1 (the impurity orbital). It is also possible to reduce the determinant in Eq. (4) to a determinant of an $N\times N$ spatial matrix,[1] but we choose not to do so here. The matrix elements of O_μ are

$$(O_\mu)_{l,l}=1, \tag{5a}$$

$$(O_\mu)_{l,l-1}=-e^{(-\Delta\tau K)}e^{V^\mu_{l-1}}(1-2\delta_{l,1}), \tag{5b}$$

and $(O_\mu)_{lm}=0$ otherwise. In Eq. (5), K is an $N\times N$ matrix corresponding to the bilinear part of the Hamiltonian [H_0 in Eq. (1)] and

$$V^\mu_l=\lambda\mu\sigma(l)|d\rangle\langle d| \tag{6}$$

is a potential acting only at the impurity site. The Green's function $g_\mu=O_\mu^{-1}$ is found to obey the Dyson equation (we omit μ indices for simplicity)

$$g'=g+(g-1)(e^{V'-V}-1)g' \tag{7}$$

relating any two spin configurations. Here $(V)_{ll'}=\delta_{ll'}V_l$ is a diagonal matrix in space and time. Equation (7) is most easily established by finding first the Dyson equation for $\tilde{g}=e^Vg$.

Because the potential acts only at the d site, Eq. (7) provides directly an $L\times L$ matrix equation for the d Green's function:

$$g'_{dd}(l,l')=g_{dd}(l,l')+[g_{dd}(l,l'')-\delta_{l,l''}][\exp(V'_{l''}-V_{l''})-1]g'_{dd}(l'',l'). \tag{8}$$

The ratio of determinants for flipping of the spin at time slice l is given by

$$R = 1 + [1 - g_{dd}(l,l)][\exp(V_l' - V_l) - 1]. \tag{9}$$

This can be shown directly by use of $\tilde{g}$. When a move is accepted, all components of the d Green's function are updated through the relation

$$g_{dd}'(l_1,l_2) = g_{dd}(l_1,l_2) + [g_{dd}(l_1,l) - \delta_{l_1,l}] \times [\exp(V_l' - V_l) - 1]\{1 + [1 - g_{dd}(l,l)][\exp(V_l' - V_l) - 1]\}^{-1} g_{dd}(l,l_2), \tag{10}$$

which is easily derived from Eq. (8). Equation (10) is formally identical to the equation derived by BSS to update the equal-time Green's function for different spatial matrix elements. This is of course obvious in the present formulation, since both follow from the Dyson equation (7).

Equations (8)–(10) constitute the basic equations of our approach. Initially, Eq. (8) is used to obtain g_{dd} from the Green's function for the fields σ set equal to zero,

$$g_{dd}^0(l,l') = \beta^{-1} \sum_n e^{-i\omega_n \Delta\tau(l-l')} g_{dd}^0(i\omega_n), \tag{11}$$

$$g_{dd}^0(i\omega_n) = -\left[i\omega_n - \left(\epsilon_d + \frac{U}{2}\right) - \sum_k \frac{|V_k|^2}{i\omega_n - \epsilon_k}\right]^{-1} \tag{12}$$

[$\omega_n = (2n+1)\pi/\beta$], by inversion of an $L \times L$ matrix (L^3 operations). After this, g_{dd} is updated in L^2 operations [Eq. (10)] every time a move is accepted. Surprisingly, this procedure is found to be very stable, and only after ~ 50 sweeps does it become necessary to recompute g from Eq. (8) at the lowest temperatures studied ($L = 128$). It should be emphasized that in this approach the conduction electrons enter the problem only through the Green's function at the impurity site, and thus it is straightforward to study different band structures or even free electrons in a continuum. The energy scale set by the bandwidth does not limit the lowest temperature that can be reached with the algorithm, as in the BSS approach.

For the single Anderson impurity we have chosen a flat density of states and an infinite bandwidth, and V_k independent of k. The starting Green's function is[4]

$$g_{dd}^0(l,l') = -\int_{-\infty}^{\infty} \frac{d\epsilon}{\pi} \frac{\Delta}{(\epsilon - \epsilon_d - U/2)^2 + \Delta^2} e^{-\epsilon\Delta\tau(l-l')}[f(\epsilon) - \theta(l-l')] \tag{13}$$

with $\Delta = \pi V^2 \rho(\epsilon_F)$, $\rho(\epsilon_F)$ the density of states at the Fermi surface, and $f(\epsilon)$ the Fermi function. We consider the symmetric case, i.e., $\epsilon_d = -U/2$. We measure observables in the usual way[1,3]; for example, the d-spin susceptibility is

$$\chi = \int_0^\beta d\tau \langle [d_\uparrow^\dagger(\tau) d_\downarrow(\tau) + d_\downarrow^\dagger(\tau) d_\uparrow(\tau)][d_\uparrow^\dagger(0) d_\downarrow(0) + d_\downarrow^\dagger(0) d_\uparrow(0)] \rangle \tag{14}$$

and the averages for each σ configuration are easily obtained from the d Green's function. The results shown here were obtained from 5000 measurement sweeps preceded by 1000 warmup sweeps; the error bars are smaller than the points except where shown. As a check on our Monte Carlo algorithm we have done extensive comparison with exact diagonalization results for an impurity interacting with a two-site conduction-electron lattice.

Figure 1 shows impurity charge and spin susceptibilities for $\Delta = 0.5$ and $U = 0$, 1, and 2. As U increases, the charge susceptibility is suppressed while the spin susceptibility increases rapidly as T decreases and levels off at low temperatures, as expected. The arrows indicate the expected low-temperature limits from the exact Bethe-*Ansatz* solution[5,6]:

$$\chi(T=0) = \chi_{\text{Kondo}}\left[1 + \frac{1}{\sqrt{\pi}} \int_0^{1/2u} \frac{dx}{\sqrt{x}} e^{x - \pi^2/16x}\right], \tag{15a}$$

$$\chi_{\text{Kondo}} = (2\pi\Delta)^{-1} (\pi/2u)^{1/2} e^{\pi^2/8u - 1/2u} \tag{15b}$$

$$\chi_{\text{ch}} = \frac{2}{\pi\Delta(2\pi u)^{1/2}} \int_{-\infty}^{\infty} dx \frac{e^{-x^2/2u}}{1 + (\pi u/2 + x)^2}, \tag{15c}$$

with $u = U/\pi\Delta$.

Figure 2 shows results for the local moment $\langle \sigma_z^2 \rangle$ and for $T\chi$ plotted versus $\log T$ for $\Delta = 0.5$ and $U = 1$,

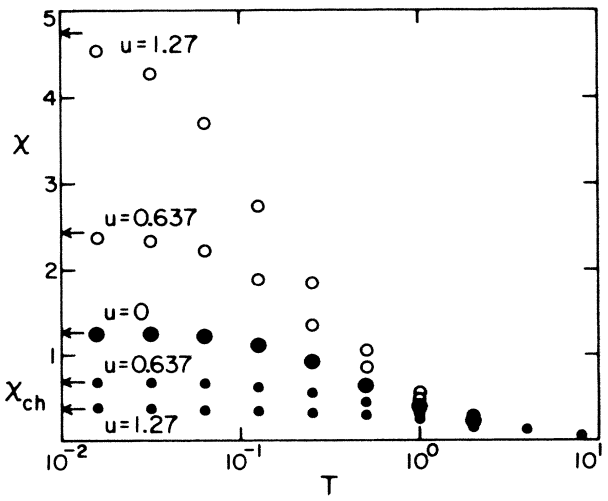

FIG. 1. Charge (closed circles) and spin (open circles) susceptibilities for a single symmetric Anderson impurity; $\Delta=0.5$ and $u=U/\pi\Delta=0$, 0.637, and 1.27. The arrows indicate the expected low-temperature limits. $\Delta\tau=0.25$.

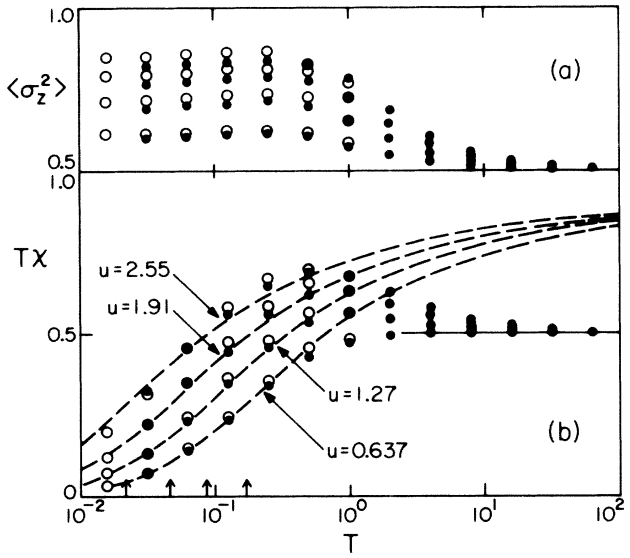

FIG. 2. (a) Local moment $\langle\sigma_z^2\rangle$ and (b) $T\times$ (spin susceptibility) for a single Anderson impurity; $\Delta=0.5$ and $u=0.637$, 1.27, 1.91, and 2.55. The closed and open circles correspond to $\Delta\tau=0.25$ and $\Delta\tau=0.5$, respectively. The dashed lines are the universal Kondo susceptibility for the four values of T_K given in the text.

2, 3, and 4. For $U=1$, there is a direct transition between the free-orbital regime ($T\chi=0.5$) and the strong-coupling regime where the local moment is quenched and $T\chi$ goes to zero. For $U\geq 2$, $T\chi$ first increases as T is lowered and the local moment starts to build up, and only at lower temperatures is the local moment quenched. The Kondo temperature obtained from the renormalization-group analysis[7] $\chi(T=0)=0.103/T_K$, with $\chi(T=0)$ obtained from Eq. (15), is $T_K=0.169$, 0.0865, 0.0435, and 0.0216 for $U=1$, 2, 3, and 4, respectively. It is shown as arrows in Fig. 2, and the corresponding curves for the universal Kondo susceptibility[7] as dashed lines. Note that the agreement with the Monte Carlo results is very good (no adjustable parameters). Our results for $U=2$ and $U=4$ compare also well to the Bethe-*Ansatz* results of Okiji and Kawakami[8] for those cases over the entire temperature range. As U becomes large, systematic errors in the Monte Carlo results due to finite $\Delta\tau$ become significant and deviations from the universal behavior occur. The error in the observables due to finite $\Delta\tau$ is expected to be proportional to $\Delta\tau^2$ and finite as $T\rightarrow 0$.[9]

It is straightforward to generalize this method to n impurities, in which case g_{dd} and V_l in Eqs. (8)–(10) become $n\times n$ matrices; the number of operations per update is $(nL)^2$, and per sweep $(nL)^3$. Figure 3 shows spin-spin correlations as a function of k_FR for two Anderson impurities with $\epsilon_d=-U/2$, $U=2$, and $\Delta=0.5$. Here, we have taken for the conduction electrons a free-electron dispersion relation:

$$\epsilon(k)=(D/2)[(k/k_F)^2-1], \quad 0\leq k\leq\sqrt{2}k_F, \quad (16)$$

and $V_{kl}=Ve^{i\mathbf{k}\cdot\mathbf{R}_l}$, $\mathbf{R}=\mathbf{R}_1-\mathbf{R}_2$. The spin-spin correlations show the expected oscillatory behavior due to the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Figure 4 shows the behavior of the susceptibility and of spin correlations versus temperature for $k_FR=0.5$, i.e., in the ferromagnetic regime. $T\chi$ is still very large around the Kondo temperature for the single impurity, indicating that the magnetic interactions dominate over the Kondo effect in this case. The staggered susceptibility is somewhat suppressed from the single-impurity value and actually levels off around T_K in this

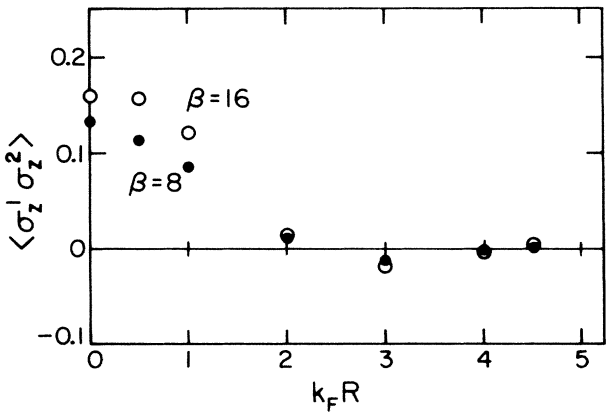

FIG. 3. Spin-spin correlations vs k_FR for two Anderson impurities; $\Delta=0.5$, $u=1.27$, $D=12$, $\Delta\tau=0.25$, and $\beta=8$ (closed circles) and 16 (open circles).

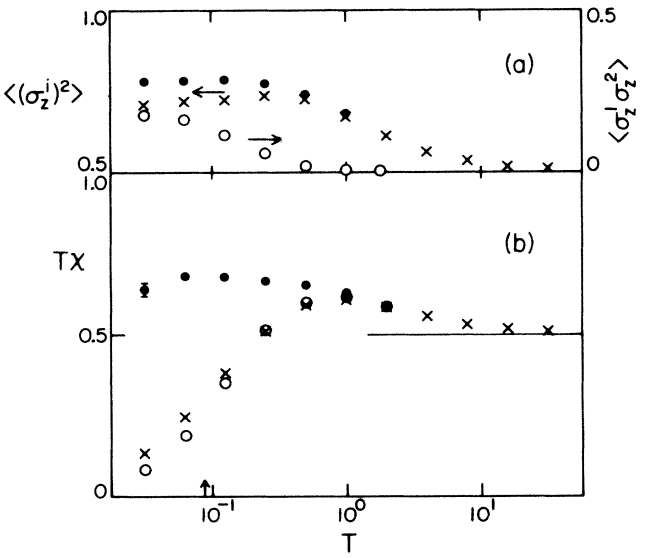

FIG. 4. (a) Spin correlations for two impurities; parameters as in Fig. 3; $k_F R = 0.5$. Open circles, $\langle \sigma_z^1 \sigma_z^2 \rangle$ (right scale); closed circles, $\langle \sigma_z^2 \rangle$ (left scale). We also show $\langle \sigma_z^2 \rangle$ for $k_F R = \infty$ (crosses). (b) $T \times$ susceptibility for $k_F R = 0.5$. Closed circles, uniform; open circles, staggered susceptibility. The crosses show the single-impurity ($k_F R = \infty$) values. The arrow indicates the Kondo temperature for the single impurity.

case. The spin-spin correlation function increases as T is lowered but appears to level off, suggesting that $\langle \sigma_1^z \sigma_2^z \rangle < 1$ at $T = 0$, so that the impurities do not become locked into a triplet state.[10] The local moment is found to be somewhat larger than for $R = \infty$. For temperatures well above T_K, the results join onto the single-impurity results.

In summary, we have discussed a method to perform simulations on magnetic impurity systems and implemented it for one and two Anderson impurities. Our approach is essentially a numerical implementation of the sum over paths in the functional integral discussed by Hamann in the single-impurity case,[4] and by Chakravarty and Hirsch for the two-impurity case[11]; one important difference, however, is that the Ising formulation allows us to rigorously sum only over the important "hopping paths" discussed by Hamann, since the other paths (small Gaussian fluctuations) are not present in this formulation; the convergence of the numerical calculation is thus greatly enhanced.[12] Our approach allows us to deal with an infinite sea of conduction electrons, and does not suffer from instabilities. For the single-impurity case, we showed that Monte Carlo simulation can reproduce the expected universal behavior; for two impurities, we showed in one case how magnetic interactions become important as the temperature is lowered. A detailed discussion of the properties of two Anderson impurities will be given elsewhere.

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