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2000 J. Phys.: Condens. Matter 12 L575

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LETTER TO THE EDITOR

Spin diffusion in the double-exchange model at intermediate temperatures

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Received 3 July 2000

Abstract. Using a combined numerical and analytic approach, we evaluate the spin-diffusion coefficient D for the double-exchange model in the limit $t \ll T \ll J_H$, where t is the hopping energy, T is the temperature, and J_H is the Hund's coupling. To lowest order, $D \propto tq(1-2q)/T\chi$ where q is given in terms of the band filling p by either p ($p < 1/2$) or $1-p$ ($p > 1/2$). Hence, the spin-diffusion coefficient vanishes when the electrons are unable to hop between singly-occupied sites in a half-filled ($p = 1/2$) band.

The subject of spin diffusion in paramagnetic systems has long been a source of fascination to condensed-matter physicists. One of the central objects in this field is the spin-diffusion coefficient (SDC) D , which determines the lifetime $\tau(k) = 1/Dk^2$ for a magnetic disturbance with wavevector k in the hydrodynamic limit of small wavevectors and frequencies [1, 2]. Correspondingly, the spin correlation function $G^{zz}(\mathbf{k}, t) = \langle S_z(\mathbf{k}, t)S_z(-\mathbf{k}, 0) \rangle$ falls off like $\exp(-t/\tau(k))$ for large times.

Even in the simplest magnetic systems of local moments interacting through Heisenberg exchange, the nature of spin diffusion has been hotly debated since the first theoretical studies were performed more than 35 years ago [3]. Deviations from the expected hydrodynamic result [1] at infinite temperature have been found [4–6] to grow as the dimension decreases. Despite the fundamental importance of these studies, the spin diffusion of itinerant systems has only been examined in the past few years. Unfortunately, recent studies of spin diffusion within the $t-J$ [7] and Hubbard [8] models were flawed by the adaptation of an approximation designed for local-moment systems but unsuitable for mobile electrons.

In [9], a new combined analytic and numerical approach was developed to study spin diffusion in itinerant systems. This technique was specifically applied to the double-exchange model, which was proposed by Zener [10] to describe the magnetic properties of the manganites [11]. The double-exchange model is characterized by a Hund's coupling J_H which tends to align the spins of the N_{el} conduction electrons with local moments at N lattice sites. In the limits $t \ll J_H \ll T$, where t is the hopping energy and T is the temperature, it was found in [9] that D is proportional to $tp(1-p)/T\chi$, where $p = N_{el}/2N$ is the band-filling fraction and χ is the magnetic susceptibility. This result implies that the SDC is proportional to the electron bandwidth and is independent, to lowest order, of J_H . The present work extends that calculation to the physically more interesting regime of intermediate temperatures, $t \ll T \ll J_H$, in which the Hund's coupling may be expected to play a prominent role. Notice that this limit still places the temperature well above the Curie temperature $T_C \sim t$, below which the itinerant and local spins become ferromagnetically ordered.

Part of the motivation for this work comes from recent experiments [12–14] which have detected diffusive dynamics in manganites such as $\text{La}_{1-y}\text{Ca}_y\text{MnO}_3$, doped with holes away

from half filling ($p = 1/2$ or one conduction electron per site) by $y = 1 - 2p \approx 0.3$ so that the magnetoresistance is near maximum. Like other manganites in this doping regime, $\text{La}_{1-y}\text{Ca}_y\text{MnO}_3$ undergoes a transition from a paramagnetic insulator above the Curie temperature T_C to a ferromagnetic metal below. It is widely believed that the double-exchange model describes the physics of this transition: above T_C , the conduction electrons are unable to readily move between sites while satisfying the local Hund's coupling due to the random orientation of the local $S = 3/2$ moments; below T_C , the long-range order of the local moments permits metallic conduction. The limit $t \ll J_H$ is expected to hold very well in manganites with $t \sim 0.2$ eV and $J_H \sim 2$ eV. In those compounds with Curie temperatures smaller than 300 K, neutron-scattering measurements [12, 13] have revealed that the spin dynamics becomes quite complex just below T_C . In addition to the expected spin-wave modes, experiments have also detected a central peak with width $\Gamma(k) = 2/\tau(k)$ proportional to k^2 , which is characteristic of spin diffusion[†]. The measured SDC $D = \Gamma/(2k^2)$ is about 15 meV \AA^2 . This diffusive peak (assuming that is what it is) grows as T approaches T_C from below and persists above T_C with little change in width.

The Hamiltonian of the double-exchange model is usually written

$$H = -t \sum_{(i,j)} \left(c_{R_i,\alpha}^\dagger c_{R_j,\alpha} + c_{R_j,\alpha}^\dagger c_{R_i,\alpha} \right) - 2J_H \sum_i \mathbf{s}_i \cdot \mathbf{S}_i \quad (1)$$

where $c_{R_i,\alpha}^\dagger$ is the creation operator for an electron with spin $\alpha = \uparrow$ or \downarrow (repeated spin indices are summed), $\mathbf{s}_i = (1/2)c_{R_i,\alpha}^\dagger \boldsymbol{\sigma}_{\alpha\beta} c_{R_i,\beta}$ is the electron spin on site i , and \mathbf{S}_i is the local spin at this site (within the spin correlation function and spin polarization, the total spin at site i is given by $\mathbf{S}_{tot,i} = \mathbf{s}_i + \mathbf{S}_i$). Since J_H is positive, electron hopping is favoured between neighbours R_i and R_j with the same orientation of the local spin. The hopping energy t is easily related to the bandwidth W by $W = 2zt$, where z is the number of nearest neighbours on the lattice. For future reference, the kinetic and potential terms in the double-exchange Hamiltonian will be denoted K and V . In order to avoid confusion with the hopping energy t , we will denote the time by x .

As shown in [9], the spin current in the direction \mathbf{a}_γ with spin polarization α is given by

$$J_\gamma^\alpha = -\frac{t}{2i} \sum_i c_{R_i,\beta}^\dagger \sigma_{\beta\kappa}^\alpha (c_{R_i+\mathbf{a}_\gamma,\kappa} - c_{R_i-\mathbf{a}_\gamma,\kappa}). \quad (2)$$

The SDC is then given by the zero-frequency moment of the spin-current correlation function (SCCF) $C(x)$ [2]:

$$\frac{\hbar D}{a^2} = \frac{t^2}{T\chi} \int_0^\infty dx C(x) \quad (3)$$

$$C(x) = \frac{1}{2N} \langle (I(x) + I(-x))I(0) \rangle \quad (4)$$

where χ is the magnetic susceptibility containing both itinerant and local-moment contributions, and $I(x) = \exp(iHx)I \exp(-iHx)$ is the time-dependent, normalized spin current with $I = \sum_i I_i = J_\gamma^z/t$.

[†] Spin diffusion produces the spin correlation function [2]

$$G^{zz}(\mathbf{k}, \omega) = \frac{2}{1 - e^{-\omega/T}} \frac{\omega D k^2 \chi}{\omega^2 + (Dk^2)^2}$$

which is characterized by a quasi-elastic peak with width $\Gamma(\mathbf{k}) = 2Dk^2$.

Earlier efforts [7, 8] to evaluate the SDC for itinerant systems adapted the Gaussian approximation [15], which was originally developed for Heisenberg systems [3]. To implement this approximation, the SCCF $C(x)$ is first expanded in powers of time as

$$C(x) = \sum_{n=0}^{\infty} \frac{(-1)^n x^{2n}}{(2n)!} C_{2n} \quad (5)$$

with $C_0 = (1/N)\langle I^2 \rangle$, $C_2 = (1/N)\langle [H, [H, I]] I \rangle$, $C_4 = (1/N)\langle [H, [H, [H, [H, I]]]] I \rangle$, and so forth. Assuming that $C(x)$ takes the Gaussian form $C(x) = a \exp(-bx^2)$, then $a = C_0$, $b = C_2/2C_0$, and only the two lowest-order coefficients are required to obtain the SDC: $\hbar D/a^2 = (t^2 C_0/2T\chi)\sqrt{2\pi C_0/C_2}$. Since $C_0 = p(1-p)$ and $C_2 \propto J_H^2$, this approximation implies that $\hbar D/a^2$ is proportional to t^2/J_H for all temperatures.

In [9], we demonstrated that the Gaussian approximation cannot be applied to itinerant systems due to the complexity of the SCCF. As in that earlier work, we again restrict consideration to a Bethe lattice with coordination number $z \gg 1$ so that closed loops are avoided and we treat the local spins classically with $S \gg 1$. Towards these ends, we define the scaled hopping and Hund's coupling constants $t = t'/\sqrt{z}$ and $J_H = J'_H/S$. In the more precisely written limits $t' < W \ll T \ll J'_H$, we may ignore the contribution of the kinetic energy to the density matrix $\rho = \exp(-\beta(V - \mu N_{el}))$, which breaks into the product of density matrices $\rho_i = \exp(-\beta(V_i - \mu N_{el,i}))$ at each site.

The calculation of the SDC now proceeds in three stages. First, we evaluate the SCCF coefficients C_n to as high an order as feasible. In the infinite temperature limit with $\rho \rightarrow \exp(\beta\mu N_{el})$, each coefficient must start with a commutator of the potential energy: terms such as

$$\begin{aligned} \langle [K, [V, [K, [V, I]]]] I \rangle &= \frac{1}{\text{Tr } \rho} \text{Tr}\{\rho [K, [V, [K, [V, I]]]] I\} \\ &= -\frac{1}{\text{Tr } \rho} \text{Tr}\{\rho [V, [K, [V, I]]] [K, I]\} \end{aligned} \quad (6)$$

vanish because $[K, \rho] = 0$ and $[K, I] = 0$. For temperatures much less than J'_H but still much greater than W , ρ no longer commutes with the kinetic energy and terms like the one above can no longer be ignored. As a result, each coefficient C_n with $n \geq 2$ contains two sets of contributions:

$$C_n = \sum_{m=0}^{n-2} \{qA(m, n-m) + q^2B(m, n-m)\} t'^m J_H'^{n-m} \quad n \geq 2 \quad (7)$$

where q is given by p for bands less than half full ($p < 1/2$) and $1-p$ for bands more than half full ($p > 1/2$). Hence, $0 \leq q \leq 1/2$ and q denotes the filling fraction of carriers (whether electrons or holes). The second set of contributions proportional to q^2 in equation (7) arises from terms such as equation (6), which do not occur at infinite temperature.

We have evaluated both sets of contributions in equation (7) to 16th order. Up to $n = 8$, the nonzero terms are $A(0, 2) = 2$, $\{A(0, 4) = 8, A(2, 2) = 4, B(2, 2) = 6\}$, $\{A(0, 6) = 32, A(2, 4) = 52, A(4, 2) = 20, B(2, 4) = 40, B(4, 2) = 50\}$, and $\{A(0, 8) = 128, A(2, 6) = 416, A(4, 4) = 462, A(6, 2) = 140, B(2, 6) = 224, B(4, 4) = 658, B(6, 2) = 448\}$. It is easy to show that to all orders, $A(0, n) = 2^{n-1}$ and $B(0, n) = 0$. So for localized electrons with $W = 0$, the normalized SCCF $C(x)/C_0$ is given by $f_0(x) \equiv 1 - \sin^2(J'_H x)/(1-q)$ with a period of π/J'_H . Although the SCCF may become negative for $q > 0$, the time-averaged SCCF $\bar{C}(x) = C_0 \bar{f}_0(x) = q(1-2q)/2$ with $t' = 0$ is always positive.

Next, we apply Tchebycheff bounds [16] to the SCCF using the fact that all Fourier components of any correlation function must be positive [2]. As shown in figure 1, the upper

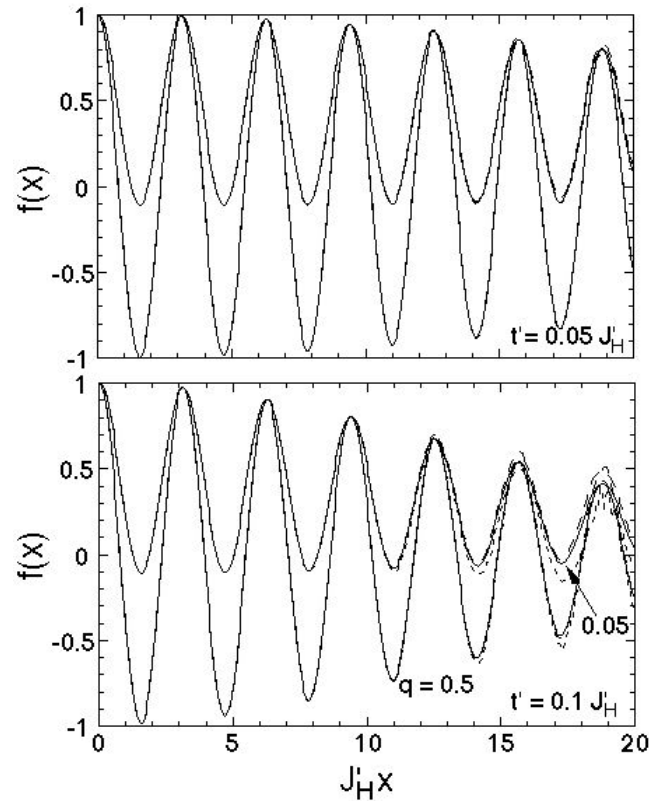


Figure 1. The dimensionless SCCF $f(x)$ versus $J'_H x$ for two values of the hopping energy t' and band filling q . The solid curve is the conjectured form of equation (8) and the dashed curves are the Tchebycheff bounds to the SCCF.

and lower Tchebycheff bounds (plotted as dashed curves) remain close together far beyond the time $x \approx 3.5/J'_H$ at which the truncated Taylor expansion blows up. The SCCF in figure 1 is plotted for two values of the band filling ($q = 0.05$ and 0.5) and for two values of the hopping energy ($t' = 0.05 J'_H$ and $0.1 J'_H$). As expected, electron hopping causes the SCCF to decay with time.

Finally, we use the Tchebycheff bounds as a guide to construct an analytic form for the SCCF. For small t'/J'_H , the scaled correlation function $f(x) = C(x)/C_0$ can be approximated by

$$f(x) = \exp(-t'^2 x^2/4) f_0(x) + (1 - \exp(-t'^2 x^2/4)) \left\{ \frac{1+q}{1-q} \left(\frac{\sin(J'_H x)}{J'_H x} \right)^2 - \frac{q}{1-q} \frac{\sin(2J'_H x)}{J'_H x} \right\} \quad (8)$$

which is plotted in the solid curves of figure 1. Notice that this expression obeys the required limits $f(0) = 1$, $\lim_{J'_H \rightarrow 0} f(x) = 1$, and $\lim_{t' \rightarrow 0} f(x) = f_0(x)$. The remaining terms in the Taylor expansion, not included in equation (8), are of order $(t'/J'_H)^4 (J'_H x)^6$ and higher. Consequently, the Tchebycheff bounds in figure 1 are weaker for $t'/J'_H = 0.1$ than for 0.05 .

After integrating the SCCF over time, we find the remarkably simple result

$$\frac{\hbar D}{a^2} = \frac{t' \sqrt{\pi}}{2zT\chi} q(1 - 2q) \quad (9)$$

with formal corrections of order $t'(t'/J'_H)^2$. Because our approach has already neglected terms of order $(t'/T)^2$ from the density matrix, we must set all higher-order corrections to zero in the limit $T \ll J'_H$. For comparison, the lowest-order term in the SDC for infinite temperature is obtained by replacing $q(1-2q)$ by $q(1-q) = p(1-p)$ in the above expression. Both results for the SDC are plotted in figure 2. The leading-order contribution to the SDC comes from the first term in equation (8), which is produced by the spin current leaving a region of short-range magnetic order. By contrast, the second term in equation (8) proportional to $1 - \exp(-t'^2 x^2/4)$ is produced by the unpolarized spin current entering this region and does not contribute to the SDC in the limit $t' \ll J'_H$.

At this point, the reason for the failure of the Gaussian approximation is clear: the second-order term C_2 used in the Gaussian approximation to construct D is independent of t' due to the cancellation between the first and second terms in the SCCF of equation (8). However, the dominant contribution to the SDC arises from the modulation of the first term in equation (8) by $\exp(-t'^2 x^2/4)$.

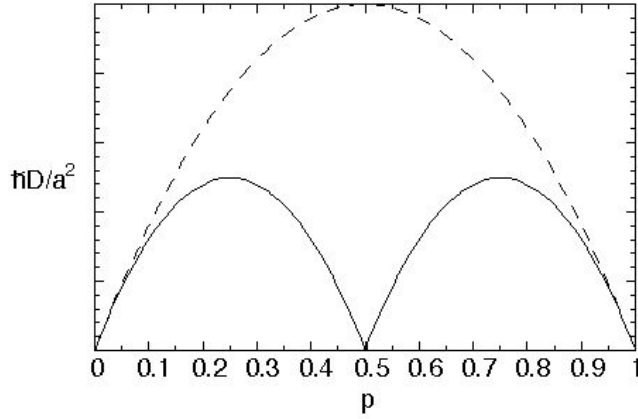


Figure 2. The SDC versus band filling p for infinite temperature (dashed) and intermediate temperatures (solid).

The most interesting effect of lowering the temperature from the high-temperature regime $T \gg J'_H \gg t'$ to the intermediate-temperature regime $J'_H \gg T \gg t'$ is that the SDC vanishes to lowest order at half filling. This effect is easy to understand. In order to maintain the Hund's coupling at half filling, every site must be occupied by a single electron. Consequently, electrons cannot hop to neighbouring, occupied sites without sacrificing enormous potential energy and the SDC must vanish. If higher-order terms in t'/J'_H or t'/T were included in this calculation, then the number of doubly-occupied and empty sites would be small but nonzero at half filling and rather than vanish, the SDC would go through a deep minimum at $p = 1/2$. Neutron-scattering measurements on a series of manganites such as $\text{La}_{1-y}\text{Ca}_y\text{MnO}_3$ doped away from $y = 0$ or $p = 1/2$ should be able to test this result: the width of the central peak far above T_C should increase linearly with y for small y (or like y^2 very close to $y = 0$ if the minimum value of D is non-negligible).

For small q , the extra set of terms $q^2 B(n, m-n)$ in the coefficient C_n can be neglected and the on-site expectation values $\langle c_{i\alpha}^\dagger c_{i\beta} \rangle = p \delta_{\alpha\beta}$ and $\langle I_i^2 \rangle = p(1-p)$ are unchanged from their values at infinite temperature. So for small numbers of electrons or holes ($q \ll 1$), the SDC is identical to its value at $T = \infty$ with $\hbar D/a^2 \approx t' \sqrt{\pi} q / 2zT\chi$. This result is somewhat counterintuitive: at infinite temperature, charge carriers can freely hop to any neighbouring

site whether or not the Hund's coupling is satisfied at that site; at intermediate temperatures, a carrier hopping from site i to j must satisfy the Hund's coupling at site j where the local spin is uncorrelated from the local spin at site i . As first shown by Anderson and Hasegawa [17], the effect of Hund's coupling in the limit $t \ll J_H$ is to introduce the effective hopping energy $t_{ij} = t \cos(\theta_{ij}/2)$, where θ_{ij} is the angle between the local spins at sites i and j . Since an electron or hole will preferentially hop to sites j with large t_{ij} , one might expect that the SDC will be suppressed at intermediate temperatures even for small carrier concentrations. But for $W \ll T$ the average kinetic energy of a charge carrier is unchanged from its value at infinite temperature and sites which are originally inaccessible to a charge carrier because $t_{ij}/t \ll 1$ may be visited by that carrier after its spin is randomized by a few hops. Consequently, the overall rate of spin diffusion approaches the same limiting value for small q .

Because our calculation was performed on a Bethe lattice, the spin dynamics follows the hydrodynamic predictions: for large times, the spin correlation function $G^{zz}(\mathbf{k}, x)$ decays like $\exp(-x/\tau(k))$ with lifetime $\tau(k) = 1/Dk^2$. Of course, we cannot rule out the possibility that there will be deviations from this result in three and lower dimensions. But studies of the Heisenberg model [6] suggest that those deviations will be minor in three dimensions. In finite dimensions, the magnitude of D may be changed by the contributions of closed loops which are not present in the Bethe lattice. The lowest-order correction from a closed loop in a hypercubic lattice adds a term of order $(J'_H)^2(t')^4 x^6/z$ to the SCCF. Since some terms of order $(J'_H)^2(t')^4 x^6$ have already been neglected in equation (8) for the SCCF, the contributions of closed loops should not significantly change our estimate for D in three dimensions for small t'/J'_H .

For a hole doping of $y = 0.3$ or a band filling of $p = 0.35$, our result for the SDC is lowered by about half from its infinite-temperature value. This implies that the calculated SDC is still roughly six times larger than its measured value in the manganites. The enhancement of the susceptibility χ near a ferromagnetic transition or corrections from the finite coordination number z and local spin S may account for most of the remaining discrepancy with the experimental results. However, our work can still not explain the coexistence of spin diffusion with long-range magnetic order below T_C [9].

To summarize, we have studied the diffusive spin dynamics of the double-exchange model using a new technique which combines Tchebycheff bounds with an analytic approximation for the SCCF. Unlike earlier methods, this new technique captures the complex behaviour of itinerant systems. In the limits $W \ll T \ll J_H S$, we find that the SDC is proportional to the electron bandwidth, is independent of the Hund's coupling, and vanishes at half filling. These results can be tested on a series of paramagnetic manganese alloys doped away from half filling by adding holes.

Helpful conversations with Drs Jaime Fernandez-Baca, Pengcheng Dai, Mike Hilliard, Stephen Lovesey, and Richard Wood are gratefully acknowledged. This research was sponsored by the US Department of Energy under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed by UT-Battelle, LLC.

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