Spin diffusion in the double-exchange model far above the Curie temperature

R S Fishman

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6032, USA

Abstract

Spin diffusion within the double-exchange model is studied in the limits $W \ll T \ll J_H S$ (intermediate temperatures) and $W \ll J_H S \ll T$ (infinite temperature), where W is the electron bandwidth, T is the temperature, S is the local spin, and J_H is the Hund's coupling. In both limits, T is still far above the Curie temperature $T_C \sim W$. All dynamical properties are obtained from the spin-current correlation function $C(x)$, where x denotes time. While $C(x)$ is real (even) at infinite temperature, it contains both real (even) and imaginary (odd) parts at intermediate temperatures. Upper and lower Tchebycheff bounds are used to evaluate the real part of $C(x)$ in each limit. From $C(\omega)$, we construct the spin conductivity $D(\omega)$, which has Gaussian peaks at $\omega = 0$ and $\pm 2 J_H S$, all with the same width $\sim W$. Whereas the central peak is produced by the hopping of electrons between sites, the side peaks are produced by the mutual precession of the local and itinerant spins at every site. At infinite temperature, each of the side peaks has half the weight of the central peak. But at intermediate temperatures, the side peaks are reduced by $T/(J_H S) \ll 1$ as the spin precession becomes energetically prohibitive. A rigorous f-sum rule relates the integral over $D(\omega)$ to the average kinetic energy at any temperature. In the zero-frequency limit, the spin-diffusion coefficient $D_s = (1/2)D(\omega = 0)$ yields the relaxation time $\tau_r(k) = 1/(D_s k^2)$ for a magnetic disturbance with wavevector k. Whereas D_s reaches a maximum at half-filling (an average of one electron per site) for infinite temperature, it vanishes at half-filling for intermediate temperatures because an electron cannot hop to a neighbouring site without sacrificing enormous Hund's energy. The predictions of this work are compared with recent neutron-scattering measurements on the manganites.

1. Introduction

Spin diffusion is the process of magnetic relaxation in a paramagnetic system [1]. A magnetic disturbance with wavevector k relaxes in time $\tau_r(k) = 1/(D_s k^2)$, where D_s is the spin-diffusion coefficient (SDC). Although well understood in local-moment systems [2], spin diffusion has

only recently been examined in itinerant systems [3–5], where a magnetic disturbance relaxes through electron hopping. This paper applies a newly developed technique [6, 7] to study the process of spin diffusion within the double-exchange model.

The Hamiltonian of the double-exchange model [8] is usually written as

$$H = -t \sum_{\langle i,j \rangle} (c^{\dagger}_{R_i \alpha} c_{R_j \alpha} + c^{\dagger}_{R_j \alpha} c_{R_i \alpha}) - 2J_{\mathrm{H}} \sum_i s_i \cdot S_i. \qquad (1)$$

Here, $c^{\dagger}_{R_i \alpha}$ is the creation operator for an electron with spin $\alpha = \uparrow$ or $\downarrow$ (repeated spin indices are summed), $s_i = (1/2) c^{\dagger}_{R_i \alpha} \vec{\sigma}_{\alpha \beta} c_{R_i \beta}$ is the spin of an electron on site i, and S_i is the local spin at that site. So the total spin at site i is given by $S_{\mathrm{tot},i} = s_i + S_i$. Since $J_{\mathrm{H}} > 0$, the Hund's coupling aligns the electron and local spins and electrons prefer to hop between neighbours R_i and R_j with parallel local spins.

Due to the close connection between electron conduction and magnetic order, the double-exchange model qualitatively explains the metal–insulator transition observed in manganites [9] like $La_{1-y}Ca_y MnO_3$ at the Curie temperature T_{C} when $y \approx 0.3$. Above T_{C}, electron conduction is impeded by the random orientation of the local moments; below T_{C}, the conductivity is enhanced by the alignment of the local moments. A metal–insulator transition can also be produced by applying a magnetic field above T_{C}, where the dramatic reduction in the resistivity is known as colossal magnetoresistance. Despite the success of the double-exchange model in explaining many qualitative features of the metal–insulator transition in the manganites, it cannot quantitatively explain the drop in resistance below T_{C} [10]. Nor can it explain many anomalous features [11, 12] of the spin dynamics below T_{C}. Rather, the effects of lattice and magnetic polarons are believed to be just as important [13] as the physics of double exchange.

The central object of this study will be the spin-current correlation function (SCCF) $C(x)$, where x is taken to be time in order to avoid confusion with the hopping energy t. Two limits will be examined: $W \ll T \ll J_{\mathrm{H}} S$ (intermediate temperatures) and $W \ll J_{\mathrm{H}} S \ll T$ (infinite temperature), where W is the bandwidth of the electrons. In both limits, T is far greater than the Curie temperature $T_{\mathrm{C}} \sim W$ [14] and short-range magnetic order is absent. Whereas the conduction-band electrons are insensitive to the effect of the Hund's coupling at infinite temperature, the electrons are restricted to states with the maximal value of $s_i \cdot S_i$ at intermediate temperatures.

In each limit, calculations are performed on a Bethe lattice with bandwidth $W = 4\sqrt{z}t$ and coordination number $z \gg 1$. Previous studies [14] of spin dynamics within the double-exchange model have also been performed on a Bethe lattice, which like a three-dimensional cubic lattice has well-defined band edges. The Bethe lattice also has the convenient property that closed loops are avoided. After some initial manipulations, we shall assume that the local spins are classical with $S \gg 1$.

Starting with the SCCF, we evaluate the spin conductivity $D(\omega)$, which is used to parametrize the neutron-scattering cross section for small k. In the limit $\omega \to 0$, $D(\omega)$ determines the SDC $D_s = D(\omega = 0)/2$. For infinite temperature, the SCCF is real and an even function of time. But for intermediate temperatures, $C(x) = C^{(1)}(x) + iC^{(2)}(x)$ contains both real (even) and imaginary (odd) contributions.

At both infinite and intermediate temperatures, the real part of the SCCF will be evaluated in three steps. First, the Taylor coefficients of $C^{(1)}(x)$ are obtained using an exact algorithm on the Bethe lattice. The Taylor coefficients (evaluated up to as high an order as possible) are then used to yield upper and lower Tchebycheff bounds for $C^{(1)}(x)$. Together, the Taylor expansion and the Tchebycheff bounds are employed to construct an analytic form for $C^{(1)}(x)$ which obeys all formal limits. This technique was previously introduced in two short publications [6, 7]

which emphasized that earlier approaches [3, 4] to spin diffusion in itinerant systems were flawed by the use of an approximation [15] designed for local-moment systems.

The spin conductivity $D(\omega)$ is then simply obtained from $C^{(1)}(\omega)$. Like $C^{(1)}(\omega)$, $D(\omega)$ has three Gaussian peaks: a central peak at $\omega = 0$ and two side peaks at $\pm 2 J_H S$, all with the same width proportional to W. Physically, the side peaks are produced by the mutual precession of the electron and local spins at any site while the central peak is produced by the hopping of electrons between sites. At infinite temperature, the weight of each side peak in $D(\omega)$ is half as large as the weight of the central peak. But at intermediate temperatures, the side peaks are suppressed by $T/(J_H S) \ll 1$ and disappear as the energy required for the spin precession becomes inaccessible. For both temperature regimes, a rigorous f-sum rule relates the expectation value of the kinetic energy K (the first term in the Hamiltonian of equation (1)) to the integral over $D(\omega)$.

Finally, we evaluate the SDC D_s from the zero-frequency limit of $D(\omega)$. When J_H/W and T/W are infinite, D_s is proportional to W/z in both temperature ranges. At infinite temperature, D_s vanishes at band-filling fractions of $p = 0$ (no electrons per site) or 1 (two electrons per site) and reaches a maximum at $p = 1/2$ (half-filling or an average of one electron per site). But for intermediate temperatures, D_s vanishes at half-filling and reaches maxima at $p = 1/4$ and $3/4$. The suppression of D_s near half-filling at intermediate temperatures has a simple significance: assuming $J_H S/T = \infty$, doubly occupied or empty sites are energetically prohibited and there is exactly one electron on each site at half-filling. Hence, the electrons are trapped at singly occupied sites and spin diffusion is prohibited.

The next section develops the basic formalism of spin diffusion. Then, section 3 summarizes the analytical and numerical technique employed to evaluate the real part of the SCCF. Results for the spin conductivity and SDC are provided in section 4. Finally, we compare those results with neutron-scattering measurements in section 5. Proof of the f-sum rule, results for the Taylor coefficients, and expressions for the Fourier transform $C^{(1)}(\omega)$ are contained in three appendices.

2. Formalism of spin diffusion

This section outlines the basic formalism [16] for spin diffusion in a paramagnet. As is well known, neutron-scattering studies measure the total spin correlation function

$$G^{\alpha\beta}(\boldsymbol{k}, \omega) = \sum_i \int \mathrm{d}x \, \mathrm{e}^{\mathrm{i}\omega x - \mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i} G^{\alpha\beta}(\boldsymbol{R}_i, x) \tag{2}$$

$$G^{\alpha\beta}(\boldsymbol{R}_i - \boldsymbol{R}_j, x) = \langle S^{\alpha}_{\mathrm{tot},i}(x) S^{\beta}_{\mathrm{tot},j}(0) \rangle. \tag{3}$$

Due to the isotropy of the spin fluctuations above T_C, $G^{\alpha\alpha}(\boldsymbol{k}, \omega)$ are all equivalent and the off-diagonal terms vanish. Hence, we shall take $\alpha = \beta = z$ and drop the spin indices. The correlation function defined above is simply related to the response function

$$\chi_2(\boldsymbol{k}, \omega) = \frac{1}{2} \sum_i \int \mathrm{d}x \, \mathrm{e}^{\mathrm{i}\omega x - \mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_i - \boldsymbol{R}_j)} \langle [S^z_{\mathrm{tot},i}(x), S^z_{\mathrm{tot},j}(0)] \rangle \tag{4}$$

through the fluctuation-dissipation theorem:

$$\chi_2(\boldsymbol{k}, \omega) = \frac{1 - \mathrm{e}^{-\beta\omega}}{2} G(\boldsymbol{k}, \omega) \tag{5}$$

where $\hbar = 1$ and $\beta = 1/T$. Of course, all time dependence is governed by the Hamiltonian H with $A(x) = \exp(\mathrm{i}Hx) A \exp(-\mathrm{i}Hx)$.

The usual hydrodynamic expression for the response function in the limit of small momentum and low frequency [1] is

$$\chi_2(\boldsymbol{k}, \omega) = \frac{\omega k^2 D_s}{\omega^2 + (D_s k^2)^2} \chi \tag{6}$$

where χ is the static susceptibility. Consequently, the correlation function contains a quasi-elastic peak with width $\Gamma(k) = 2/\tau_r(k) = 2D_s k^2$. We shall employ a somewhat more general relation [16] which is valid for small k but arbitrary ω:

$$\chi_2(\boldsymbol{k}, \omega) = \operatorname{Im} \chi(\boldsymbol{k}, z = \omega + i\epsilon) \tag{7}$$

$$\chi(\boldsymbol{k}, z) = \frac{i k^2 \bar{D}(z)}{z + i k^2 \bar{D}(z)} \chi \tag{8}$$

$$\bar{D}(z) = \int_0^\infty \mathrm{d}x \, e^{izx} D(x) = \int \frac{\mathrm{d}\omega}{2\pi i} \frac{D(\omega)}{\omega - z} \qquad \operatorname{Im} z > 0 \tag{9}$$

where $\epsilon > 0$ is an infinitesimal. This parametrization of the response function is valid for $ka \ll 1$ and $k\xi \ll 1$, where ξ is the magnetic correlation length. So for a fixed k, it will fail in the critical regime very close to T_C, when ξ becomes large. For z in the upper half-plane, equation (9) implies that

$$\bar{D}(\omega + i\epsilon) = -i \, \mathrm{P} \int \frac{\mathrm{d}\omega'}{2\pi} \frac{D(\omega')}{\omega' - \omega} + \frac{1}{2} D(\omega) \tag{10}$$

where P is the principal part of the integral. We shall refer to the imaginary part of $\bar{D}(\omega + i\epsilon)$ as $-i \, \mathrm{P} \, I$.

It follows that the spin conductivity can be written

$$D(\omega) = \frac{2\omega}{\chi} \lim_{k \to 0} \frac{1}{k^2} \chi_2(\boldsymbol{k}, \omega) = \frac{\omega}{\chi} (1 - e^{-\beta\omega}) \lim_{k \to 0} \frac{1}{k^2} G(\boldsymbol{k}, \omega). \tag{11}$$

But the right-hand side is related to the SCCF through [16]

$$\lim_{k \to 0} \frac{1}{k^2} G(\boldsymbol{k}, \omega) = \frac{a^2 t^2}{\omega^2} C(\omega) \tag{12}$$

$$t^2 C(x) = \lim_{k \to 0} \frac{1}{N} \langle J_\gamma^z(\boldsymbol{k}, x) J_\gamma^z(-\boldsymbol{k}, 0) \rangle \tag{13}$$

where γ is fixed and J_γ^α is the spin current in the direction $\boldsymbol{a}_\gamma$ (which lies along one of $z/2$ mutually orthogonal directions) with polarization α. Hence, $D(\omega)$ and $C(\omega)$ are connected by

$$D(\omega) = \frac{a^2 t^2}{\chi} \frac{1 - e^{-\beta\omega}}{\omega} C(\omega) \tag{14}$$

and the SDC $D_s = D(\omega = 0)/2$ is obtained by integrating $C(x)$ over all time:

$$D_s = \frac{a^2 t^2}{2\chi T} C(\omega = 0) = \frac{a^2 t^2}{2\chi T} \int \mathrm{d}x \, C(x). \tag{15}$$

Another useful quantity is the 'memory time' τ_m, defined by

$$\tau_m = \frac{1}{D(x = 0)} \int_0^\infty \mathrm{d}x \, D(x) = \frac{D_s}{D(x = 0)} \tag{16}$$

which yields the range of $D(x)$ in time. Unlike the relaxation time $\tau_r(k)$, τ_m corresponds to a microscopic collision time [16].

To calculate the spin current, we apply the equation of continuity for the total spin containing *both* local and itinerant contributions:

$$i\frac{dS^{\alpha}_{tot,k}}{dt} = [S^{\alpha}_{tot,k}, H] = \sum_{\delta} k \cdot a_{\delta} J^{\alpha}_{\delta}(k) \tag{17}$$

$$J^{\alpha}_{\gamma}(k) = \frac{t}{2i} \sum_{i} e^{-ik \cdot R_i} c^{\dagger}_{R_i,\beta} \sigma^{\alpha}_{\beta\kappa} (c_{R_i+a_{\gamma},\kappa} - c_{R_i-a_{\gamma},\kappa}) \tag{18}$$

which uses the spin-commutation relations between the quantum local spins S_i to ensure that the potential V conserves the total spin or that $[S_{tot,i}, V] = 0$. If only the itinerant spin were used in the definition of the spin correlation function $\chi_2(k, \omega)$ [5], then the commutator $[s_i, V]$ would contain the term $2i J_H s_i \times S_i$ corresponding to the precession of the itinerant and local spins. This precession term is avoided when the spin correlation function is correctly defined in terms of the *total* spin. Having included the local spin in the time derivative of $S_{tot,k}$, we find that it does not affect the spin current, which only involves Fermion operators[1]. At this point (but not before), it is safe to take S to be large and to treat the local spins classically. Exploiting the isotropy above T_C once again, we define the dimensionless spin current $I(x) = J^z_{\gamma}(k = 0)/t$ so that the dimensionless SCCF is given by $C(x) = (1/N)\langle I(x)I(0)\rangle$, independent of γ.

Since all Fourier components of the correlation function $C(x)$ are positive [16], the real and imaginary parts of $C(x) = C^{(1)}(x) + iC^{(2)}(x)$ must be even and odd functions of x, respectively. It follows from equation (15) that D_s is real and proportional to the integral of $C^{(1)}(x)$. Furthermore, $C(\omega) = C^{(1)}(\omega) + C^{(2)}(\omega) > 0$ also contains two contributions:

$$C^{(1)}(\omega) = \int dx \cos(\omega x) C^{(1)}(x) \tag{19}$$

$$C^{(2)}(\omega) = -\int dx \sin(\omega x) C^{(2)}(x) \tag{20}$$

where $C^{(1)}(\omega)$ is an even and $C^{(2)}(\omega)$ is an odd function of ω. The next section introduces a technique for evaluating $C^{(1)}(x)$ and $C^{(1)}(\omega)$. Both $C^{(2)}(x)$ and $C^{(2)}(\omega)$ may then be obtained from the symmetry relation $C(-\omega) = \exp(-\beta\omega)C(\omega)$, which implies that

$$C^{(2)}(\omega) = \tanh(\beta\omega/2)C^{(1)}(\omega). \tag{21}$$

Combining this expression with equation (14) yields the spin conductivity:

$$D(\omega) = \frac{2a^2 t^2}{\chi} \frac{\tanh(\beta\omega/2)}{\omega} C^{(1)}(\omega) \tag{22}$$

which is always a positive and even function of ω, as required by the condition $\omega\chi_2(k, \omega) \geqslant 0$.

Completing this formal development, the f-sum rule derived in appendix A relates the integral over $D(\omega)$ to the average kinetic energy and to the memory time for any itinerant model where the potential conserves the total spin:

$$\int \frac{d\omega}{2\pi} D(\omega) = -\frac{a^2}{2Nz\chi}\langle K\rangle = \frac{D_s}{\tau_m}. \tag{23}$$

Unlike the results obtained below for $C(\omega)$ and $D(\omega)$, this sum rule is completely general and is not restricted to temperatures far above T_C or to the Bethe lattice. Nor does it make any assumptions about the sizes of S and z.

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[1] This corrects a sign mistake made for the spin current in earlier papers [6,7]. Since the SCCF involves the product of two spin currents, this sign error had no deleterious effects.

3. Tchebycheff bounds and the spin-current correlation function

This section outlines a calculation of the real SCCF $C^{(1)}(x)$ within the double-exchange model. For simplicity, this calculation is performed on a Bethe lattice with $z \gg 1$ and for classical local spins with $S \gg 1$. We shall work with the scaled parameters $t' = t\sqrt{z} = W/4$ and $J'_H = J_H S$. As discussed in earlier publications [6, 7], the calculation is performed in three steps.

First, $C^{(1)}(x)$ is expanded in a power series:

$$C^{(1)}(x) = \sum_{n=0}^{\infty} \frac{(-1)^n x^{2n}}{(2n)!} C_{2n} \tag{24}$$

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. In the absence of electron hopping with $t' = 0$, the coefficients C_{2n} may be evaluated exactly from the commutators of the potential V with the spin current I. The real SCCF is then given by $C^{(1)}(x) = p(1-p)\cos^2(J'_H x)$ (infinite T) or $C^{(1)}(x) = q(1-q) - q\sin^2(J'_H x)$ (intermediate T), where p is the filling fraction of electrons ($p = 1$ corresponds to two electrons per site so the single band is filled) and q is the filling fraction of *carriers*. Hence, q equals p for $p < 1/2$ (the carriers being electrons) or $1 - p$ for $p > 1/2$ (the carriers being holes). In either case, q lies between 0 and 1/2. Notice that $C^{(1)}(x = 0) = C_0 = p(1-p) = q(1-q)$ is the same in both ranges of temperature. The oscillation of $C^{(1)}(x)$ with frequency $2J_H S$ is caused by the mutual precession of the electron and local spins at any site, during which the total spin remains constant.

When $t' > 0$, we must evaluate all possible commutators of $H = K + V$ with the spin current I. Each coefficient C_{2n} can be evaluated numerically but exactly using the commutation relations between V, K, and I. The contributions to each coefficient are parametrized as

$$C_{2n} = p(1-p) \sum_{m=0}^{n-1} A(2m, 2n-2m)(t')^{2m}(J'_H)^{2n-2m} \qquad \text{infinite } T \tag{25}$$

$$C_{2n} = \sum_{m=0}^{n-1} \{q A(2m, 2n-2m) + q^2 B(2m, 2n-2m)\}(t')^{2m}(J'_H)^{2n-2m} \qquad \text{intermediate } T \tag{26}$$

with $n \geqslant 1$. Results for $A(2m, 2n-2m)$ and $B(2m, 2n-2m)$ are tabulated in appendix B up to 16th order.

Since $C(\omega) = C^{(1)}(\omega) + C^{(2)}(\omega) > 0$, where $C^{(1)}(\omega)$ is an even function of frequency while $C^{(2)}(\omega)$ is odd, it follows that $C^{(1)}(\omega)$ must also be positive for all ω. Using this result together with the constraints $d^{2n}C^{(1)}(x)/dx^{2n}|_{x=0} = (-1)^n C_{2n}$, we obtain upper and lower Tchebycheff bounds [17] to $C^{(1)}(x)$. Unlike the truncated Taylor expansion, which blows up for relatively small values of $J'_H x$, the Tchebycheff bounds remain close together up to much longer times. Results for these bounds are provided in [6] and [7].

Finally, the Tchebycheff bounds and the Taylor coefficients are used to construct analytic forms for $C^{(1)}(x)$. These forms are required to satisfy the limits $C^{(1)}(x = 0) = p(1-p)$ and $\lim_{J'_H \to 0} C^{(1)}(x) = p(1-p)$. Of course, $C^{(1)}(x)$ must also reduce to the exact forms given above when $t = 0$. For small t'/J'_H, the real SCCFs are well approximated by the expressions

$$C^{(1)}(x) = p(1-p)\left\{ \exp(-t'^2 x^2/4) \frac{1 + \cos(2J'_H x)}{2} \right.$$
$$\left. + (1 - \exp(-t'^2 x^2/4)) \frac{1 - \cos(2J'_H x)}{2(J'_H x)^2} \right\} \qquad \text{infinite } T \tag{27}$$

$$C^{(1)}(x) = \frac{q}{2} \exp(-t'^2 x^2/4)(1 - 2q + \cos(2J'_{\mathrm{H}}x)) + q(1 - \exp(-t'^2 x^2/4))$$
$$\times \left\{ (1+q)\frac{1 - \cos(2J'_{\mathrm{H}}x)}{2(J'_{\mathrm{H}}x)^2} - q\frac{\sin(2J'_{\mathrm{H}}x)}{J'_{\mathrm{H}}x} \right\} \qquad \text{intermediate } T. \qquad (28)$$

Due to the spin current flowing out of the region R containing the local magnetic disturbance, electron hopping damps the dominant first term by $\exp(-t'^2 x^2/4)$. The decay of the outgoing spin polarization as it enters a region outside R with randomized local spins produces the second terms in equations (27) and (28), which are proportional[2] to $1 - \exp(-t'^2 x^2/4)$. Since this electron polarization decays by precessing about the randomized local spins outside R, those terms fall off with a $1/x^2$ or $1/x$ dependence. Hence, the SCCF decays with two very different timescales: a Gaussian decay in time $1/t'$ and a power-law decay in time $1/J'_{\mathrm{H}}$. In the limit $t' \ll J'_{\mathrm{H}}$, the randomization of the electronic spin polarization outside R occurs very quickly and the slow Gaussian decay of the magnetic fluctuation inside R dominates the magnetic response. For both temperature regimes, equation (15) implies that the leading-order term in D_s is proportional to t and is independent of J_{H}.

Earlier approaches [3–5] to spin diffusion in itinerant models adapted a technique from local-moment systems by assuming that $C^{(1)}(x)$ takes the purely Gaussian form $C^{(1)}(x) = a \exp(-bx^2)$ [15]. Then $a = C_0$ and $b = C_2/2C_0$ are simply obtained from the first two Taylor coefficients and the SDC is given by $D_s = (a^2 t^2 C_0/2T\chi)\sqrt{2\pi C_0/C_2}$. Since $C_0 = p(1-p)$ and $C_2 \propto J_{\mathrm{H}}^2$, this approximation implies that D_s is proportional to t^2/J_{H} for all temperatures!

For the double-exchange model, the Gaussian approximation fails because the lowest-order $t'^2 x^2$ terms from the two exponentials in equation (27) or (28) cancel one another. Therefore, the Gaussian approximation cannot correctly identify the time dependence of the outgoing spin current, which dominates the process of spin diffusion for small t'/J'_{H}. On a more physical level, the Gaussian approximation fails because spin diffusion in the double-exchange model (and more generally, in any itinerant model) is a two-step process: first, the electron spin polarization leaves region R of the magnetic disturbance; then, this electronic spin polarization decays by interacting with the randomized spins outside R. Due to the separate contribution of each step, the SCCF cannot be approximated as a Gaussian.

Previous applications of the Tchebycheff method were restricted to local-moment systems. For a Heisenberg ferromagnet, the SCCF involves the product of four spin operators S_i^z so it is easier to deal directly with the spin correlation function $G(k, \omega)$. Several groups [18–20] have developed sophisticated techniques for applying Tchebycheff bounds to $G(k, x)$ in time. Unlike for a Fermion system, however, each commutation with the Hamiltonian increases the number of spin operators by one. So it is very cumbersome to evaluate expectation values like $\langle [H, [H, \ldots [H, S_i^z] \ldots]] S_j^z \rangle$ to high order. Consequently, study has been restricted to isotropic and XXZ, spin-1/2 Heisenberg chains at infinite temperature.

4. Spin conductivity

This section uses the results obtained in the previous section for $C^{(1)}(x)$ together with the formalism developed in section 2 to obtain the full SCCF $C(x) = C^{(1)}(x) + iC^{(2)}(x)$ as well as the Fourier transforms $C(\omega)$ and $D(\omega)$. Those results then provide the SDC D_s and the memory time τ_m. The f-sum rule introduced earlier is used to evaluate the average kinetic energy in both temperature regimes. The two temperature regimes are considered separately.

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[2] This paper provides a somewhat different (i.e. more mature) explanation of the $1 - \exp(-t'^2 x^2/4)$ term in the SCCF than in earlier publications [6,7].

4.1. Infinite temperature

It is straightforward but tedious to evaluate the Fourier transform of equation (27). The full result for $C^{(1)}(\omega)$ in equation (C.1) has peaks at $\omega = 0$ and $\pm 2 J'_H$, the central peak produced by electron hopping and the side peaks produced by the precession of the electron and local spins. To leading order in t'/J'_H, $C^{(1)}(\omega)$ can be written in the much simpler form

$$C^{(1)}(\omega) = p(1-p)\frac{\sqrt{\pi}}{t'}\left\{e^{-\omega^2/t'^2} + \frac{1}{2}\left(e^{-(\omega-2J'_H)^2/t'^2} + e^{-(\omega+2J'_H)^2/t'^2}\right)\right\}. \quad (29)$$

Notice that each of the three Gaussian peaks in $C^{(1)}(\omega)$ has an identical width proportional to t' or W and that the weight of each side peak is half the weight of the central peak. By contrast, the Gaussian approximation assumes that $C^{(1)}(\omega)$ contains only a single Gaussian peak centred at $\omega = 0$. In the limit $t' \to 0$, $(\sqrt{\pi}/t')\exp(-\omega^2/t'^2) \to \pi\delta(\omega)$ and $C^{(1)}(\omega)$ is the superposition of three delta functions.

Since T is much larger than any characteristic frequency of $C^{(1)}(\omega)$, equation (21) indicates that $C^{(2)}(\omega)$ must vanish. Hence, $C^{(2)}(x) = 0$ and $C(x) = C^{(1)}(x)$ is purely real. Results for $C(x)$ are plotted in figure 1(a) for $p = 0.35$ and $t'/J'_H = 0.05$ or 0.10. As expected, the SCCF $C(x)$ falls off much more rapidly for the higher value of the hopping energy.

The spin conductivity can now be obtained from equation (22):

$$D(\omega) = \frac{a^2 t'^2}{T\chi z}C(\omega) \quad (30)$$

with the same proportionality holding between $D(x)$ and $C(x)$. Normalized by $D(\omega = 0) = 2D_s$, $D(\omega)$ is plotted in figure 2(a). As expected from equation (29), $D(\omega)$ has two side peaks with a combined weight equal to the central peak. The imaginary part of $\bar{D}(\omega + i\epsilon)$ is given by equation (C.3). For small ω/t', this result reduces to

$$-i\,\mathrm{P}\,I \approx 2i\frac{D_s\omega}{\sqrt{\pi}t'} \quad (31)$$

which is a linear function of ω.

From equations (29) and (30), we obtain the SDC:

$$D_s = p(1-p)\frac{\sqrt{\pi}a^2 t'}{2T\chi z} \quad (32)$$

with corrections of order $t'(t'/J'_H)^2$. So to lowest order, D_s is proportional to $W/(zT\chi)$ and is independent of the Hund's coupling. Also to leading order, the memory time defined by equation (16) is given by $\tau_m = \sqrt{\pi}/(2t')$, independent of the band-filling fraction. Hence, the range of $D(x)$ grows as the bandwidth decreases or, equivalently, as the Fermi velocity increases.

Together with the f-sum rule of equation (23), our result for $D(x)$ implies that the average kinetic energy is given by

$$\langle K\rangle = -\frac{2t'^2}{T}Np(1-p) \quad (33)$$

which can also be obtained directly from the double-exchange Hamiltonian. Notice that $\langle K\rangle$ vanishes for an empty or full band but reaches a minimum at half-filling with $p = 1/2$. Of course, doubly occupied and empty sites are energetically allowed at half-filling when $T \gg J_H S$.

Due to the absence of correlation between the local and itinerant spins, the magnetic susceptibility at infinite temperature is given by

$$T\chi = \frac{1}{3}\langle(\mathbf{s}_1 + \mathbf{S}_1)^2\rangle = \frac{1}{2}p(1-p) + \frac{1}{3}S(S+1) \quad (34)$$

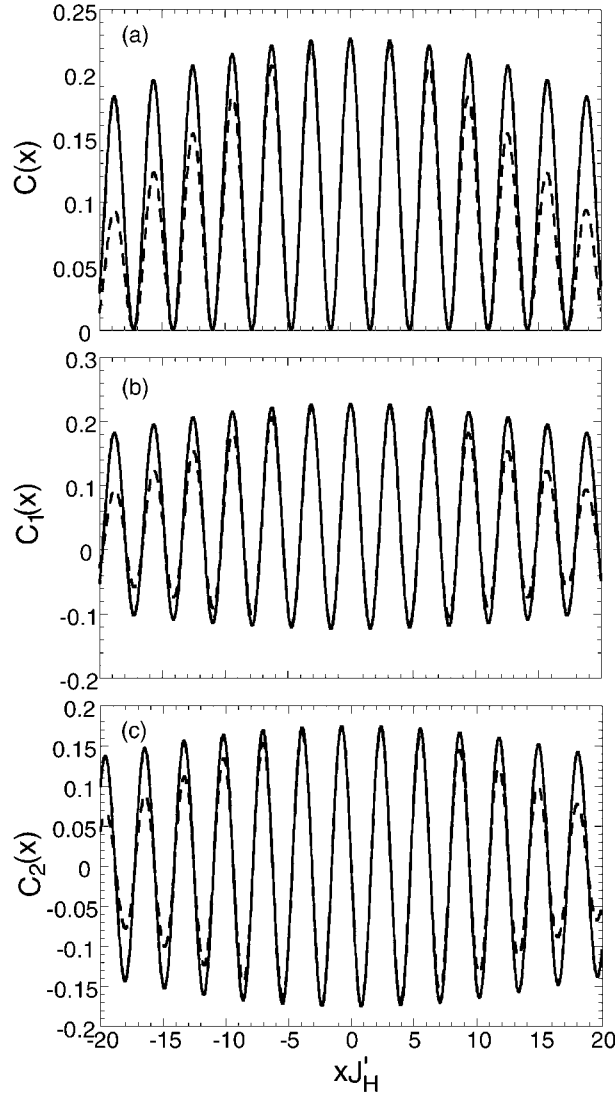with separate electronic and local contributions.

Figure 1. The time-dependent SCCF $C(x)$ versus xJ'_H in the limit of (a) infinite temperature (with $C(x)$ purely real) and intermediate temperatures (with the (b) real and (c) imaginary parts of $C(x) = C^{(1)}(x) + iC^{(2)}(x)$ plotted separately) for $q = 0.35$ and $t'/J'_H = 0.05$ (solid) or 0.1 (dashed).

4.2. Intermediate temperatures

At intermediate temperatures, the Fourier transform of equation (28) is given by equation (C.2). Just as at infinite temperature, $C^{(1)}(\omega)$ has peaks at $\omega = 0$ and $\pm 2J'_H$. To leading order in t'/J'_H, $C^{(1)}(\omega)$ can be written in the more compact form

$$C^{(1)}(\omega) = \frac{q\sqrt{\pi}}{t'} \left\{ (1 - 2q)e^{-\omega^2/t'^2} + \frac{1}{2}\left(e^{-(\omega - 2J'_H)^2/t'^2} + e^{-(\omega + 2J'_H)^2/t'^2}\right) \right\}. \quad (35)$$
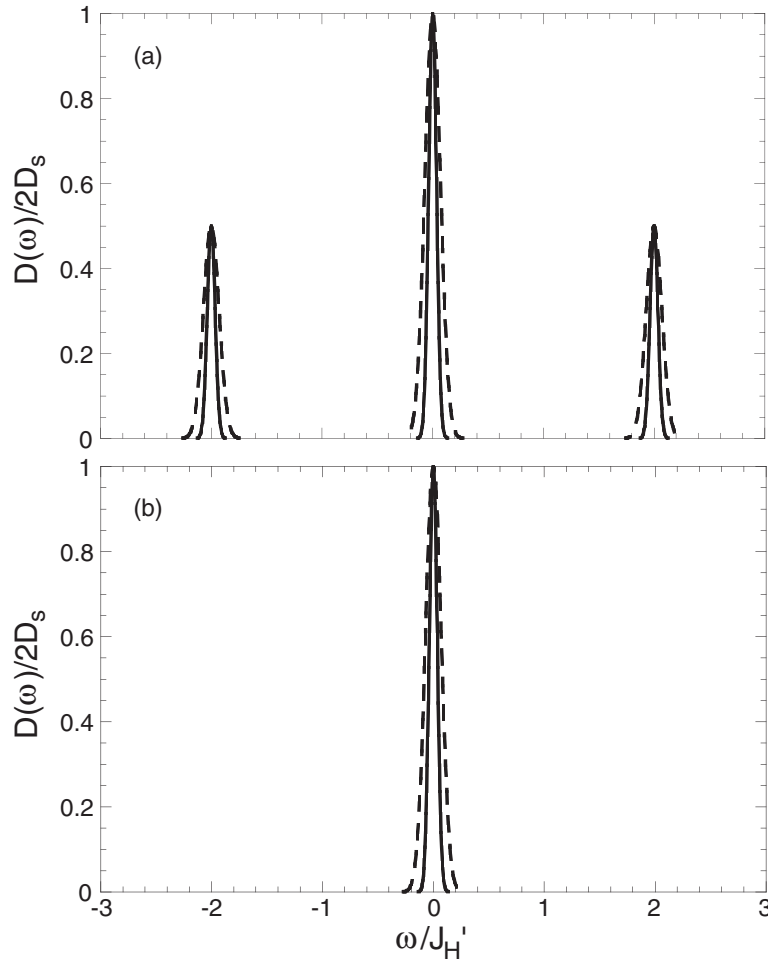
Figure 2. The spin conductivity $D(\omega)$ normalized by $2D_s$ versus ω/J'_H in the limit of (a) infinite and (b) intermediate temperature for the same parameters as in figure 1.

Once again, the presence of side peaks in $C^{(1)}(\omega)$ indicates that the Gaussian approximation fails at intermediate temperatures. From equation (21), it follows that the odd contribution to $C(\omega) = C^{(1)}(\omega) + C^{(2)}(\omega)$ is

$$C^{(2)}(\omega) = \frac{q\sqrt{\pi}}{2t'}\left\{e^{-(\omega-2J'_H)^2/t'^2} - e^{-(\omega+2J'_H)^2/t'^2}\right\}\tag{36}$$

which exactly cancels the $\omega = -2J'_H$ peak in $C^{(1)}(\omega)$.

Using the full expression for $C^{(1)}(\omega)$ in appendix C together with equation (21), we obtain the imaginary and odd part of the SCCF $C(x) = C^{(1)}(x) + iC^{(2)}(x)$:

$$C^{(2)}(x) = -\frac{q}{2}\exp(-t'^2x^2/4)\sin(2J'_Hx)$$
$$+ q(1 - \exp(-t'^2x^2/4))\left\{(1+q)\frac{\sin(2J'_Hx)}{2(J'_Hx)^2} - q\frac{\cos(2J'_Hx)}{J'_Hx}\right\}.\tag{37}$$

Since the inequality $J'_H \gg t'$ was assumed in deriving this relation, the limit $J'_H \to 0$ cannot be taken without first taking $t' \to 0$. Results for $C^{(1)}(x)$ and $C^{(2)}(x)$ are plotted in figures 1(b) and (c) for the same parameters as in figure 1(a). Both the real and imaginary parts of $C(x)$ are damped exponentially by electron hopping.

To leading order in t'/J'_H, equation (22) implies that the spin conductivity can be written as

$$D(\omega) = \frac{q\sqrt{\pi}a^2 t'}{zT\chi} \left\{ (1 - 2q)e^{-\omega^2/t'^2} + \frac{T}{2J'_H} \left(e^{-(\omega-2J'_H)^2/t'^2} + e^{-(\omega+2J'_H)^2/t'^2} \right) \right\}. \tag{38}$$

Unlike the case of infinite temperature, where the weight of each side peak in $D(\omega)$ is half that of the central peak, the weight of each side peak is now suppressed by an amount proportional to T/J'_H because the frequency of the spin precession is far above T. Neglecting the small side peaks and normalizing by $2D_s$, $D(\omega)$ is plotted in figure 2(b).

The imaginary part of $\bar{D}(\omega + i\epsilon)$ is provided by equation (C.4). When $\omega/t' \ll 1$, $-i\,P\,I \approx 2iD_s\omega/(\sqrt{\pi}t')$ is identical to the expression at infinite T except that the SDC is now given by

$$D_s = q(1 - 2q)\frac{\sqrt{\pi}a^2 t'}{2T\chi z} \tag{39}$$

with corrections of order $t'(t'/J'_H)^2$ and $t'(t'/T)^2$. For a small filling fraction of carriers (small $q = p$ or $1 - p$), this expression agrees with equation (32) for D_s at infinite temperature. But the new result for D_s vanishes at half-filling due to the inability of the electrons to hop to neighbouring sites without sacrificing enormous Hund's energy. So at half-filling, each site is singly occupied and the relaxation time $\tau_r(k) = 1/(D_s k^2)$ diverges. Due to the same physics, the Curie temperature of the double-exchange model also vanishes at half-filling [14].

From equation (16), the memory time at intermediate temperatures is $\tau_m = \sqrt{\pi}/t'$, twice as large as for infinite temperature. Hence, the range of $D(x)$ increases as the temperature decreases. Because it is related to the microscopic time between electronic collisions, the memory time is not affected by the band insulator which forms at half-filling. Ignoring the small side peaks in $D(\omega) \approx 2D_s \exp(-(\omega\tau_m)^2/\pi)$, we find that $D(x) \approx (D_s/\tau_m)\exp(-\pi(x/2\tau_m)^2)$. So unlike $C(x)$, $D(x)$ can be approximated as a Gaussian at intermediate temperatures with timescale τ_m. This suggests that simple approximations for $D(x)$ stand a better chance of success for itinerant models than do similar approximations for $C(x)$ [16].

Combined with the f-sum rule of equation (23), these results require that the average kinetic energy at intermediate temperatures satisfies

$$\langle K \rangle = -N\frac{t'^2}{T}q(1 - 2q) - N\frac{t'^2}{J'_H}q. \tag{40}$$

At half-filling, the first term vanishes and $-\langle K \rangle$ is suppressed by a factor of T/J'_H compared to its value for small q. In the limit $T/J'_H \to 0$, $\langle K \rangle/W$ vanishes at half-filling because the electrons are trapped at singly occupied sites. But for $T/J'_H > 0$, some double occupancy is energetically allowed and the average kinetic energy remains non-zero at half-filling.

Accounting for the correlation between the local and itinerant spins, the previous result for the magnetic susceptibility is slightly altered:

$$T\chi = \tfrac{1}{3}\langle(s_1 + S_1)^2\rangle = \tfrac{1}{2}p(1 - p) + \tfrac{1}{3}S(S + 1) + \tfrac{2}{3}Sq \tag{41}$$

where the last term arises[3] from $(2/3)\langle s_1 \cdot S_1 \rangle = 2Sq/3$. Hence, $T\chi$ is larger than at infinite temperature.

4.3. Common features in the two temperature regimes

A few conclusions can be drawn from the common features of spin diffusion in the two temperature regimes. Using the results for $\bar{D}(\omega + i\epsilon)$, we find that $\chi_2(\boldsymbol{k}, \omega)$ takes a Lorentzian form (expected in the hydrodynamic limit) provided that $\omega \ll t'$ and $k^2 D_s \ll t'$. Since the inequality $ka \ll 1$ was already assumed in order to allow neglect of the momentum dependence of $\bar{D}(z)$, the second inequality above is always satisfied.

When $ka \ll 1$ and $xt' \gg 1$, $G(\boldsymbol{k}, x)$ falls off exponentially with

$$G(\boldsymbol{k}, x) \approx T\chi e^{-D_s k^2 x} \tag{42}$$

which obeys the required limit $\lim_{k \to 0} G(\boldsymbol{k}, x) = T\chi$. Notice that the exponential time decay of the spin correlation function can also be written as $\exp(-x/\tau_r(k))$, which is the expected hydrodynamic result. Just as for the Heisenberg model [21–23], however, deviations from this behaviour might be expected as the dimension or coordination number are reduced.

This raises the question of whether our results would be qualitatively changed by using a hypercubic lattice in $d = z/2$ dimensions rather than a Bethe lattice. It has long been recognized [24] that the Bethe lattice has pathologies which become especially irksome when evaluating order parameters and critical parameters [25]. But whereas the bandwidth $W = 2zt$ of a hypercubic lattice diverges as $z \to \infty$ and $t = t'/\sqrt{z} \to 0$, the bandwidth $W = 4\sqrt{z}t$ of a Bethe lattice remains finite in this limit. Hence, the physics of the Bethe lattice in infinite dimensions may better reflect the physics of a three-dimensional system with well-defined band edges [14, 26].

The pathologies of the Bethe lattice originate from its property that the surface-to-volume ratio for large z is proportional to $1 - 1/z$ [24], which compromises the thermodynamic limit. However, in evaluating the coefficients $A(2m, 2n - 2m)$ and $B(2m, 2n - 2m)$ on the Bethe lattice, surface effects are irrelevant. To lowest order, the contribution of closed loops on a hypercubic lattice would change the expansion of $C^{(1)}(x)$ by a term proportional to $(xJ'_H)^6(t'/J'_H)^4$. For $t'/J'_H \ll 1$, terms of this order are quite small. In fact, the analytic expressions for $C^{(1)}(x)$ already neglect selected terms of this order, which were found [6] not to appreciably change the results for D_s. So it seems quite reasonable that spin relaxations on Bethe and hypercubic lattices behave in a similar fashion.

Indeed, very recent work [27] using dynamical mean-field theory (which becomes exact in infinite dimension) indicates that the spin conductivity $D(\omega)$ depends solely on the value of the density of states at the Fermi level $\rho(0)$ rather than on the lattice topology. The small difference between the values of $\rho(0)$ in the Bethe and hypercubic lattices leads to small and insignificant differences between the results for $D(\omega)$ in those two systems.

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[3] The expectation value of $V_i = -2J_H s_i \cdot S_i$ at intermediate temperatures is evaluated from the quantum-mechanical result that $s_i \cdot S_i = S/2$ ($2S + 2$ states) or $-(S + 1)/2$ ($2S$ states). So for large S,

$$\langle s_i \cdot S_i \rangle = \frac{e^{\beta\mu}}{1 + e^{\beta(\mu + J'_H)} + e^{2\beta\mu}} S \tanh(\beta J'_H) = Sq$$

where the last equality uses

$$p = \frac{1}{2} \frac{e^{\beta(\mu + J'_H)} + 2e^{2\beta\mu}}{1 + e^{\beta(\mu + J'_H)} + e^{2\beta\mu}}$$

to evaluate $\exp(\beta\mu)$ in the limit $\beta J'_H \to \infty$ for $p < 1/2$ ($\mu < 0$) or $p > 1/2$ ($\mu > 0$).
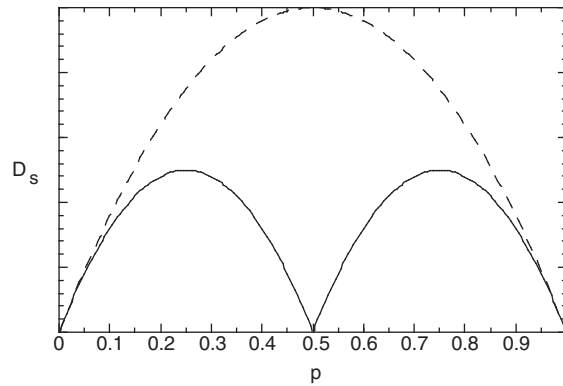
Figure 3. The SDC versus doping for infinite (dashed) and intermediate (solid) temperatures, assuming that $T\chi$ is the same in the two temperature regimes.

5. Discussion and conclusions

This paper has presented a general technique for studying spin diffusion in itinerant systems. Spin relaxation in an itinerant system occurs in two steps: first, the electronic spin current leaves region R of the magnetic disturbance; second, this outgoing electronic polarization relaxes by coupling with the randomized spins outside R. Due to the complexity of this two-step process, techniques borrowed from local-moment systems are not adequate for studying spin relaxation in itinerant systems.

The physics of the double-exchange model at temperatures far above and far below the Hund's coupling $J_H S$ is quite different: whereas the electron and local spins are uncorrelated for $T \gg J_H S$, they are highly correlated for $T \ll J_H S$. Viewed in terms of $D(\omega)$, the change in spin dynamics from infinite to intermediate temperatures is surprisingly simple with the spin precession peaks at $\omega = \pm 2 J_H S$ suppressed by $T/(J_H S)$ at intermediate temperatures. For both temperature regimes, the integral over $D(\omega)$ is proportional to the average kinetic energy and is inversely proportional to the magnetic susceptibility (more about this in a moment).

In infinite dimensions, it can be shown [27] that the spin conductivity $D(\omega)$ is proportional to the optical conductivity $\sigma(\omega)$ (see appendix A). Although derived under very general conditions, the f-sum rule for $D(\omega)$ reduces in infinite dimensions to the optical sum rule for $\sigma(\omega)$. The peak in $D(\omega)$ at $2 J_H S$ is then associated with the peak predicted by dynamical mean-field theory [28] in $\sigma(\omega)$ at the same frequency. Within the optical conductivity, the peak at $2 J_H S$ is produced by electronic transitions between spin states that are parallel and antiparallel to the local spin. Optical measurements [29] on the manganites may have detected this peak.

Results for the SDC are summarized in figure 3, which assumes that $T\chi$ is independent of temperature. In both temperature regimes, the leading-order term in D_s is proportional to the electron bandwidth W and is independent of J_H. But the doping dependences of D_s are quite different in the two regimes. While D_s has a maximum at half-filling for infinite temperature, it vanishes at half-filling for intermediate temperatures due to the prohibition of doubly occupied and empty sites. Close to $p = 1/2$ at intermediate temperatures, D_s is predicted to be a linear function of doping.

Neutron-scattering measurements [11, 12] on manganites like $La_y Ca_{1-y} MnO_3$ with hole doping $y \approx 0.3$ (and $p = (1-y)/2 \approx 0.35$) are, in at least two ways, difficult to reconcile with the predictions of the double-exchange model for spin relaxation. First, the SDC exhibits little temperature dependence as T approaches T_C [12]. Second, spin diffusion seems to persist even below T_C [11, 12], where the quasi-elastic peak in $\chi_2(\mathbf{k}, \omega)$ coexists with spin-wave peaks.

Can such behaviour arise from an exact treatment of the double-exchange model in three dimensions? Unlike our other results, the f-sum rule of equation (23) remains valid even close to T_C and in any dimension. As $T \to T_C$, $\chi \to \infty$ and the integral over $D(\omega)$ must vanish. Since $D(\omega) \geqslant 0$ for all ω, this implies that $D(\omega) \to 0$ for every ω and, in particular, that $D_s = D(\omega = 0)/2 \to 0$ as $T \to T_C$. So away from the critical regime where the magnetic correlation length ξ becomes large and our parametrization of the response function fails, the double-exchange model predicts that D_s should exhibit dramatic temperature dependence. This prediction cannot be reconciled with the measurements described above. But aside from narrowing the electron bandwidth, it is unknown how the presence of lattice and magnetic polarons may alter the spin dynamics of the double-exchange model. It is possible that the inhomogeneities produced by polarons [13] open other avenues of magnetic relaxation close to T_C.

Despite the inadequacy of the double-exchange model, it is interesting to quantitatively compare our theoretical predictions with measurements [11, 12] of D_s close to T_C. For manganites with $z = 6$ neighbours, a local spin of $S = 3/2$, and a bandwidth of $W \approx 2$ eV, we expect that $t' = W/4 \approx 0.5$ eV, $J'_H = J_H\sqrt{S(S+1)} \approx 3.9$ eV, and $t'/J'_H \approx 0.13$. For an electron filling of $p = 1/3$ corresponding to a doping of $y = 1/3$, the double-exchange model predicts that $D_s \approx 180$ meV Å^2 (infinite T) or 72 meV Å^2 (intermediate T), which corrects for the slightly larger value of $T\chi$ at intermediate temperatures. Hence, the predicted value of D_s for intermediate temperatures is only about 5 times larger than the value $D_s \approx 15$ meV Å^2 measured near T_C in the La$_{1-y}$Ca$_y$MnO$_3$ compound with $y = 0.3$ [11, 12]. Moreover, recent measurements by Dai *et al* [30] indicate that the doping dependence of D_s obeys the prediction $D_s \propto y$ of the double-exchange model for small y or for p close to $1/2$.

The relatively good agreement between the predictions of the double-exchange model at intermediate temperatures and the measurements close to T_C suggests that the double-exchange model may be a good starting point for describing spin diffusion in the manganites. Rigorous results like the f-sum rule derived in this paper may provide the framework for understanding the physics of highly complex systems like the manganites, including the non-trivial effects of polarons.

Acknowledgments

Appendix A. f-sum rule

On the basis of the definitions of $\chi_2(\boldsymbol{k}, \omega)$ in equation (4) and $J^z(\boldsymbol{k})$ in equation (18), we find that

$$\int \frac{d\omega}{\pi} \omega \chi_2(\boldsymbol{k}, \omega) = \frac{1}{N} \sum_\delta \boldsymbol{k} \cdot \boldsymbol{a}_\delta \langle [J^z_\delta(\boldsymbol{k}), S^z_{\text{tot},-\boldsymbol{k}}] \rangle. \tag{A.1}$$

But the commutator of the spin current with the total spin is given by

$$[J^z_\delta(\boldsymbol{k}), S^z_{\text{tot},-\boldsymbol{k}}] = \frac{t}{4} \boldsymbol{k} \cdot \boldsymbol{a}_\delta \sum_i \left\{ c^\dagger_{\boldsymbol{R}_i,\alpha} c_{\boldsymbol{R}_i + \boldsymbol{a}_\delta,\alpha} + c^\dagger_{\boldsymbol{R}_i,\alpha} c_{\boldsymbol{R}_i - \boldsymbol{a}_\delta,\alpha} \right\} \tag{A.2}$$

and so

$$\frac{1}{\chi} \int \frac{d\omega}{\pi} \omega \chi_2(\boldsymbol{k}, \omega) = k^2 \int \frac{d\omega}{2\pi} D(\omega) = -\frac{(ka)^2}{2Nz\chi} \langle K \rangle = \frac{k^2 D_s}{\tau_m} \tag{A.3}$$

where the first equality follows from equation (11). This f-sum rule relates the integral over $D(\omega)$ to the expectation value of the kinetic energy for any Hamiltonian with a potential V

that commutes with the total spin [27]. It is valid for all temperatures and for all values of S and z, in any dimension.

In infinite dimensions, it may be shown [27] that the spin conductivity $D(\omega)$ is simply related to the particle conductivity $\sigma(\omega)$ by

$$D(\omega) = \frac{1}{2\chi e^2}\sigma(\omega). \tag{A.4}$$

Hence, the f-sum rule derived above for $D(\omega)$ in any dimension is equivalent to the f-sum rule (also known as the optical sum rule) for the particle conductivity [31] in infinite dimensions.

Appendix B. Taylor coefficients

Here we summarize our results for the Taylor coefficients C_{2n}, which are parametrized by equations (25) and (26). Up to 16th order, $A(2m, 2n-2m)$ and $B(2m, 2n-2m)$ are given by

$A(2m2n-2m)$ m	0	1	2	3	4	5	6	7
n								
1	2							
2	8	4						
3	32	52	20					
4	128	416	462	140				
5	512	2 752	5 820	4 788	1 176			
6	2 048	16 384	55 792	85 140	54 252	11 088		
7	8 192	91 136	454 272	1 098 240	1 271 270	651 222	113 256	
8	32 768	483 328	3 312 640	11 589 760	20 965 438	19 173 440	8 138 702	1 226 940

$B(2m2n-2m)$ m	1	2	3	4	5	6	7
n							
2	6						
3	40	50					
4	224	658	448				
5	1 152	6 000	9 480	4 368			
6	5 632	45 408	125 312	132 858	45 540		
7	26 624	306 176	1 292 928	2 367 508	1 852 994	500 214	
8	122 880	1 907 200	11 386 880	31 611 554	42 323 970	25 903 306	5 725 720

Appendix C. Expressions for $C^{(1)}(\omega)$ and P I

Performing the Fourier transforms of equations (27) and (28), we find that

$$C^{(1)}(\omega) = p(1-p)\left\{\frac{\sqrt{\pi}}{t'}\left\{e^{-w^2} + \frac{1}{2}\left(e^{-(w-u)^2} + e^{-(w+u)^2}\right)\right\}\right.$$
$$+ \frac{\pi t'}{2J_{\mathrm{H}}'^2}\left\{(u-|w|)\Theta(u^2-w^2) + w\Phi(w) + \frac{1}{\sqrt{\pi}}e^{-w^2} - \frac{1}{2}(w+u)\Phi(w+u)\right.$$
$$\left.\left. - \frac{1}{2}(w-u)\Phi(w-u) - \frac{1}{2\sqrt{\pi}}\left(e^{-(w+u)^2} + e^{-(w-u)^2}\right)\right\}\right\} \qquad \text{infinite } T \quad \text{(C.1)}$$

$$C^{(1)}(\omega) = \frac{q\sqrt{\pi}}{t'}\left\{(1-2q)e^{-w^2} + \frac{1}{2}\left(e^{-(w-u)^2} + e^{-(w+u)^2}\right)\right\}$$
$$+ \frac{\pi t'}{2J_{\mathrm{H}}'^2}q(1+q)\left\{(u-|w|)\Theta(u^2-w^2) + w\Phi(w) + \frac{1}{\sqrt{\pi}}e^{-w^2}\right.$$
$$\left. - \frac{1}{2}(w+u)\Phi(w+u) - \frac{1}{2}(w-u)\Phi(w-u) - \frac{1}{2\sqrt{\pi}}\left(e^{-(w+u)^2} - e^{-(w-u)^2}\right)\right\}$$
$$- \frac{\pi}{2J_{\mathrm{H}}'}q^2\left\{2\Theta(u^2-w^2) - \Phi(w+u) + \Phi(w-u)\right\} \qquad \text{intermediate } T \quad \text{(C.2)}$$

where $w = \omega/t'$, $u = 2J'_{\mathrm{H}}/t'$, $\Theta(w)$ is the step function, and $\Phi(w)$ is the probability function ($\Phi(-w) = -\Phi(w)$ and $\Phi(w) \to 1$ as $w \to \infty$).

The imaginary part of $\bar{D}(\omega + \mathrm{i}\epsilon)$ is given by

$$-\mathrm{i}\,\mathrm{P}\,I = \mathrm{i}\frac{a^2 t'}{T\chi z}p(1-p)\left\{F(w) + \frac{1}{2}\big(F(w-u) + F(w+u)\big)\right\} \qquad \text{infinite } T \qquad (\text{C.3})$$

$$-\mathrm{i}\,\mathrm{P}\,I = \mathrm{i}\frac{a^2 t'}{T\chi z}q\left\{(1-2q)F(w) + \frac{T}{2J'_{\mathrm{H}}}\big(F(w-u) + F(w+u)\big)\right\} \qquad \text{intermediate } T$$

$$(\text{C.4})$$

$$F(w) = \mathrm{e}^{-w^2}\int_0^w \mathrm{d}u\,\mathrm{e}^{-u^2} = \frac{\pi}{2\mathrm{i}}\mathrm{e}^{-w^2}\Phi(\mathrm{i}w). \qquad (\text{C.5})$$

It can be shown that $F(w) \to w$ for $w \ll 1$ and $F(w) \to 1/(2w)$ for $w \gg 1$.

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