

## Spin diffusion of the $t$ - $J$ model

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The spin-diffusion constant of the two-dimensional  $t$ - $J$  model is calculated using an analytical approach at high temperatures and a recently developed numerical method based on the Lanczos technique combined with random sampling in the intermediate temperature regime. A simple relation,  $\sigma = D_s \chi$ , between spin conductivity and spin diffusion is established and used to calculate the latter. In the high-temperature and low-doping limit, the calculated diffusion constant agrees with known results for the Heisenberg model. At small hole doping,  $D_s$  increases approximately linearly with doping, which leads us to an important conclusion that hopping processes enhance spin diffusion at high temperatures. At modest hole doping,  $\delta \sim 0.25$ , diffusion exhibits a nonmonotonic temperature dependence, which indicates anomalous spin dynamics at small frequencies.

### I. INTRODUCTION

There appears to be a rising interest in the spin dynamics of high-temperature superconductor compounds. The research in this field is mostly concentrated around the spin-lattice-relaxation rate,  $1/T_1$ . In particular, recent measurements of  $1/T_1$  in Sr-doped  $\text{La}_2\text{CuO}_4$  (Ref. 1) display rather unusual temperature and doping dependences. Sokol *et al.*<sup>2</sup> have calculated  $1/T_1$  for the undoped case and found good agreement with experiments. They also calculated the spin-diffusion constant, which gives a short-wavelength contribution to the nuclear-relaxation rate at high temperatures. The relative strength of this diffusive contribution to the relaxation rate depends strongly on the hyperfine structure, which differs in the case of Cu and O nuclei.<sup>3,4</sup> Spin diffusion can serve also as a general numerical test probe for the spin dynamics of the system at small frequencies and momenta.

The spin-diffusion constant of a paramagnet in the limit of high temperature was first calculated by de Gennes using a moment expansion of the spin-susceptibility function.<sup>5</sup> The method was later improved by Bennett and Martin<sup>6</sup> using an exact spectral representation of the spin-correlation function in the hydrodynamic limit. Morita<sup>7</sup> investigated a theoretical criterion for the occurrence of spin diffusion by introducing a friction function. He also calculated diffusion constants for various lattices and values of spin.

In this paper we present a finite-temperature calculation of the spin-diffusion constant in the  $t$ - $J$  model on a square lattice

$$H = -t \sum_{\langle i,j \rangle, s} (c_{i,s}^\dagger c_{j,s} + \text{H.c.}) + J \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - n_i n_j / 4), \quad (1.1)$$

where  $\langle i,j \rangle$  represents summation over pairs of nearest neighbors,  $c_{i,s}^\dagger$  and  $c_{i,s}$  are creation and annihilation fermion operators restricted to the basis of single-fermion occupation number, and  $\mathbf{S}_i$  is a spin-1/2 operator.

To obtain finite-temperature static and dynamic properties of the  $t$ - $J$  model, we use a recently developed numerical method, based on the Lanczos exact-diagonalization technique combined with random sampling.<sup>8</sup> First we present an alternative method of calculating spin diffusion, which is based on the spin-conductivity function. Next, we present an analytical high-temperature result for  $D_s$  in two limits of the  $t$ - $J$  model: (a)  $t = 0$  and (b)  $J = 0$ . In the case of zero hole doping,  $\delta = 0$ , our analytical method reproduces the known result for  $D_s$  of the Heisenberg model. Next, we briefly discuss the finite-temperature Lanczos method, and then present results for  $D_s$  in the  $t$ - $J$  model at arbitrary parameter values. Last, we present conclusions.

### II. METHOD

#### A. Spin-diffusion constant

We start by noting that the total  $z$  component of the spin operator  $S^z = \sum_{\mathbf{r}} S^z(\mathbf{r})$  is a constant of motion from which follows the operator-continuity relation

$$\frac{\partial S^z(\mathbf{r}, t)}{\partial t} + \nabla \cdot \mathbf{j}_s(\mathbf{r}, t) = 0, \quad (2.1)$$

where  $\mathbf{j}_s$  is the spin- $z$ -current operator. Now, we suppose that at finite temperatures the hydrodynamic description can be applied to spin relaxation in the  $t$ - $J$  model in which the net flow of magnetization from the region of large  $S^z$  towards small  $S^z$  is governed by the diffusion equation

$$\frac{\partial \langle S^z(\mathbf{r}, t) \rangle_{\text{noneq}}}{\partial t} - D_s \nabla^2 \langle S^z(\mathbf{r}, t) \rangle_{\text{noneq}} = 0. \quad (2.2)$$

It is important to notice that contrary to Eq. (2.1), which is an operator equation, the diffusion equation is valid only with brackets  $\langle \rangle_{\text{noneq}}$ , which represent nonequilibrium ensemble averages.

Applying linear response, hydrodynamics and the fluctuation-dissipation theorem one can obtain the following form of the Fourier transform of the spin-correlation function  $S(\mathbf{r}, t) = \langle S^z(\mathbf{r}, t) S^z(0, 0) \rangle$ ;<sup>9</sup>

$$S(\mathbf{k}, \omega) = \langle S^z(\mathbf{k}, \omega) S^z(-\mathbf{k}, -\omega) \rangle \approx \frac{2}{1 - e^{-\beta\omega}} \times \frac{\omega D_s k^2 \chi}{\omega^2 + (D_s k^2)^2}, \quad (2.3)$$

valid for small  $k$  and  $\omega$ . The  $S^z(\mathbf{k}, \omega)$  is a spatial and temporal Fourier transform of  $S^z(\mathbf{r}, t)$ . We use units in which  $\hbar = k_B = 1$ . In Eq. (2.3)  $\beta$  is the inverse temperature,  $\chi$  is the static magnetic susceptibility, and  $D_s$  is the spin diffusion constant. Note that the last factor in Eq. (2.3) is the imaginary part of the dynamic susceptibility function in the hydrodynamic limit. The brackets  $\langle \rangle$  in Eq. (2.3) and all the following equations represent thermodynamic equilibrium averages. According to Onsager, the spontaneous equilibrium fluctuations of the spin, described by the spin-correlation function  $S(\mathbf{k}, \omega)$ , obey the same diffusion equation as do the nonequilibrium-induced spin fluctuations in Eq. (2.2).

Next we define the spin conductivity as

$$\begin{aligned} \sigma_x(\mathbf{k}, \omega) &= \frac{1 - e^{-\beta\omega}}{2\omega} \langle j_x(\mathbf{k}, \omega) j_x(-\mathbf{k}, \omega) \rangle \\ &= \frac{1 - e^{-\beta\omega}}{2\omega} \int_{-\infty}^{\infty} dt \sum_{\mathbf{r}} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \\ &\quad \times \langle j_x(\mathbf{r}, t) j_x(0, 0) \rangle, \end{aligned} \quad (2.4)$$

where  $j_x(\mathbf{r}, t)$  is the  $x$  component of the spin current and  $j_x(\mathbf{k}, \omega)$  its Fourier transform. The spin conductivity  $\sigma(\mathbf{k}, \omega)$  can be related to the spin response function  $S(\mathbf{k}, \omega)$  using the continuity relation (2.1) in  $(\mathbf{k}, \omega)$  space:  $-\omega S^z(\mathbf{k}, \omega) + \mathbf{k} \cdot \mathbf{j}(\mathbf{k}, \omega) = 0$ :

$$\sigma_x(\mathbf{k}, \omega) = \frac{1}{2} \omega (1 - e^{-\beta\omega}) \frac{S(\mathbf{k}, \omega)}{k^2}. \quad (2.5)$$

Combining Eqs. (2.3) and (2.5) we derive the following simple Einstein relation between the spin conductivity and spin-diffusion constant:

$$\sigma = \sigma_x(\mathbf{k} = 0, \omega = 0) = D_s \chi. \quad (2.6)$$

### B. D<sub>s</sub> in the high-temperature limit

In the high-temperature limit of the Heisenberg model it is reasonable to expect that the spin conductivity,  $\sigma(\omega)$  at  $\mathbf{k} = 0$ , can be approximated with a Gaussian form.<sup>6</sup> We have checked this assumption numerically. However,

this is in general no longer true in the case of the  $t$ - $J$  model, predominantly due to the coexistence of two different energy scales. There are two limiting cases of the  $t$ - $J$  model, however, for which we expect that a Gaussian form is still a reasonably good ansatz: (a)  $J = 1$ ,  $t = 0$ ,  $\delta \ll 1$  and (b)  $J = 0$ ,  $t = 1$ ,  $\delta \ll 1$ , where  $\delta$  represents hole doping. Note that the above assumption must be limited to small hole-doping regime. In the high-doping regime, the spin conductivity has a Drude-like (Lorentzian) form.

While a direct computation of  $\sigma(\omega = 0)$  would be subject to all the usual vagaries of numerical computation, we can avoid these [after taking the assumption of the Gaussian form of  $\sigma(\omega)$ ] by taking a ratio [see (2.10)] of the first two nonzero moments of the spin conductivity:

$$\begin{aligned} \langle \omega^0 \rangle &= \int_{-\infty}^{\infty} d\omega \sigma(\omega) = \frac{\pi}{T} \sum_{\mathbf{r}} \langle j_x(\mathbf{r}, 0) j_x(0, 0) \rangle, \\ \langle \omega^2 \rangle &= \int_{-\infty}^{\infty} d\omega \omega^2 \sigma(\omega) \\ &= \frac{\pi}{T} \sum_{\mathbf{r}} \left\langle \left( i \frac{\partial}{\partial t} \right) j_x(\mathbf{r}, t) \left( -i \frac{\partial}{\partial t} \right) j_x(0, t) \right\rangle, \end{aligned} \quad (2.7)$$

where the time derivatives are to be taken at  $t = 0$  and the index  $\mathbf{r}$  runs over all lattice sites. The spin-current operator  $j_x(\mathbf{r})$  for the  $t$ - $J$  model can be calculated using the continuity relation (2.1) together with the relation  $\frac{\partial}{\partial t} S^z(\mathbf{r}, t) = -i[S^z(\mathbf{r}, t), H]$ . Taking into account the fact that all the operators are now defined on a discrete lattice, we obtain

$$\begin{aligned} j_x(\mathbf{r}) &= -\frac{i}{2} \left[ J (S_{\mathbf{r}}^+ S_{\mathbf{r}+\hat{x}}^- - \text{H.c.}) \right. \\ &\quad \left. - t (c_{\mathbf{r}\uparrow}^\dagger c_{\mathbf{r}+\hat{x}\uparrow} - c_{\mathbf{r}\downarrow}^\dagger c_{\mathbf{r}+\hat{x}\downarrow} - \text{H.c.}) \right], \end{aligned} \quad (2.8)$$

with  $\hat{x}$  being the unit vector in the  $x$  direction. To avoid confusion we note again that  $j_x$  represents the  $x$  component of the spin- $z$ -current operator. In order to calculate the first two moments (2.7), one has to compute also the time derivative of the spin current. Taking the high-temperature expectation values we arrive after a somewhat tedious but straightforward calculation at the following expressions for the first two moments:

$$\begin{aligned} \langle \omega^0 \rangle &= \left( \frac{t^2}{2} \delta (1 - \delta) + \frac{J^2}{8} (1 - \delta)^2 \right) \frac{\pi}{T}, \\ \langle \omega^2 \rangle &= \left( \frac{t^4}{2} \delta (1 - \delta)^2 (9 + \delta) + \frac{J^4}{16} (1 - \delta)^2 (5 - 3\delta) \right. \\ &\quad \left. + O(t^2 J^2) \right) \frac{\pi}{T}. \end{aligned} \quad (2.9)$$

In the above equation, the second moment  $\langle \omega^2 \rangle$  is correct only in the two limits of interest, i.e., at (a)  $J = 0$  or (b)  $t = 0$ . Assuming a Gaussian functional form for the spin conductivity, we can express the diffusion constant in Eq. (2.6) in terms of the first two moments (2.7)

$$D_s = \frac{1}{\sqrt{2\pi}} \frac{1}{\chi} \sqrt{\frac{\langle \omega^0 \rangle^3}{\langle \omega^2 \rangle}}, \quad (2.10)$$

which leads to the final expressions for the spin-diffusion constant for the two limiting cases:

$$D_s = \begin{cases} \frac{J(1-\delta)}{2} \sqrt{\frac{\pi}{5-3\delta}} & ; \text{ for } t = 0, \\ t\delta \sqrt{\frac{2\pi}{(9+\delta)(1-\delta)}} & ; \text{ for } J = 0. \end{cases} \quad (2.11)$$

To test the method, we compare the diffusion constant (2.11) in the Heisenberg limit ( $t = 0$ ,  $\delta = 0$ ) with the known result for the Heisenberg model at high temperatures  $D_s/J = \sqrt{\frac{\pi}{20}}^{2,6}$  and find exact agreement. At  $t = 0$  and small hole doping  $\delta$ ,  $D_s$  decreases linearly with doping as  $D_s/J \sim \sqrt{\frac{\pi}{20}}(1 - \frac{7}{10}\delta)$ . This is simply related to the fact that the number of spin-current carriers (spins) decreases with doping. In the opposite limit, when  $J = 0$ , spin diffusion (2.11) increases linearly with doping as  $D_s/t \sim (2\sqrt{10}/3)\sqrt{\frac{\pi}{20}}\delta$ . It is straightforward to check that at zero doping and  $J = 0$ ,  $D_s/t = 0$ , since the spin current (2.8) at  $\delta = 0$  is zero. The linear increase of  $D_s$  with doping  $\delta$  can in this case be explained by the linear increase of spin-current carriers (holes). Interestingly, the coefficient of the linear increase of  $D_s$  in the  $J = 0$  limit is larger than the coefficient of decrease in the  $t = 0$  limit. Since the physical parameter region (relevant for cuprates), where  $J/t \sim 0.3$ , is not accessible by the present analytic method, we make the following assumption: neglecting the combined hopping and spin-flip contributions to the diffusion constant we predict that in the high-temperature regime and at  $J/t = 0.3$ ,  $D_s$  increases linearly with doping.

### C. Numerical method

Having exhausted the possibilities of analytical calculation we now turn to the numerical calculation of  $D_s$  in physically relevant parameter regimes. We used a finite-temperature Lanczos method, recently developed by one of the present authors (J.J.) in cooperation with Prelovšek. Since the details of the method have been published,<sup>8</sup> we only briefly summarize the main steps.

Taking into account the translational symmetry of the system, the spin conductivity (2.4) at  $\mathbf{k} = 0$  can be written as

$$\sigma(\omega) = \frac{1 - e^{-\beta\omega}}{2N\omega} \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle J_x(t) J_x(0) \rangle, \quad (2.12)$$

$$J_x(t) = \sum_{\mathbf{r}} j_x(\mathbf{r}, t),$$

where  $N$  is the number of sites. According to the finite-temperature Lanczos method,<sup>8</sup> the thermodynamic equilibrium average in Eq. (2.12) is expressed in terms of functions  $|\psi_m^n\rangle$ ,  $|\tilde{\psi}_k^n\rangle$  and corresponding energies  $\epsilon_{nm}$ ,  $\tilde{\epsilon}_{nk}$ , generated by the Lanczos method on a finite cluster with  $N$  sites,

$$\langle J_x(t) J_x(0) \rangle = Z^{-1} \sum_n \sum_{m,k} \langle n | \psi_m^n \rangle e^{-\beta\epsilon_{nm}} \times e^{i(\epsilon_{nm} - \tilde{\epsilon}_{nk})t} \langle \psi_m^n | J_x | \tilde{\psi}_k^n \rangle \langle \tilde{\psi}_k^n | J_x | n \rangle,$$

$$Z = \sum_n \sum_m |\langle n | \psi_m^n \rangle|^2 e^{-\beta\epsilon_{nm}}, \quad (2.13)$$

where states  $|n\rangle$  for  $n = 1$  to  $N_{\text{st}}$  represent a complete orthonormal basis set of the Hamiltonian  $H$ . In the first summation, only  $N_0 \ll N_{\text{st}}$  of states  $|n\rangle$  are randomly chosen for partial random sampling of basis states. The functions  $|\psi_m^n\rangle$  and energies  $\epsilon_{nm}$  are obtained as eigenvectors and eigenvalues of the tridiagonal form of the Hamiltonian, generated by the Lanczos procedure,

$$H|\phi_m^n\rangle = b_{nm}|\phi_{m-1}^n\rangle + a_{nm}|\phi_m^n\rangle + b_{n(m+1)}|\phi_{m+1}^n\rangle, \\ m = 0, \dots, M \ll N_{\text{st}}; \quad b_{n0} = b_{n(M+1)} = 0, \quad (2.14)$$

starting from the initial wave function  $|\phi_0^n\rangle = |n\rangle$ . Similarly,  $|\tilde{\psi}_k^n\rangle$  and  $\tilde{\epsilon}_{nk}$  are generated by the Lanczos procedure, analogous to (2.14), but with a different initial wave function  $|\tilde{\phi}_0^n\rangle = J_x|n\rangle/\sqrt{\langle n | J_x^2 | n \rangle}$ .

The static magnetic susceptibility  $\chi = \beta S(\mathbf{k} = 0)$  is evaluated with the same method, adapted to the calculation of static quantities. Here, the equal-time spin correlation function is expressed as

$$S(\mathbf{k}) = Z^{-1} \sum_n \sum_m e^{-\beta\epsilon_{nm}} \langle n | \psi_m^n \rangle \langle \psi_m^n | S^z(\mathbf{k}) S^z(-\mathbf{k}) | n \rangle, \quad (2.15)$$

where  $S^z(\mathbf{k}) = N^{-1/2} \sum_{\mathbf{r}} e^{-i\mathbf{k}\cdot\mathbf{r}} S^z(\mathbf{r})$ , and the rest of notation is the same as in Eq. (2.13).

The method yields very accurate results even for a severely reduced number of Lanczos steps  $M \ll N_{\text{st}}$  and number of random states  $N_0 \ll N_{\text{st}}$ . Provided that  $N_0$  is large enough,  $\sigma_x(\omega)$  has correct frequency moments  $\langle \omega^p \rangle$  in the limit  $T \rightarrow \infty$  for  $p = 0, \dots, M$  (for a finite cluster). Similarly, at finite temperatures the corresponding double series for the moments of  $\sigma(\omega)$  are correct up to  $\beta^{p+1} \langle \omega^{M-p} \rangle$ , for  $p = 0, \dots, M$ . The method also gives reliable results at  $T = 0$ , assuming that the number of Lanczos steps  $M$  is larger than the number of Lanczos steps required to obtain the corresponding ground-state results. For a typical run,  $M = 100$ – $150$  Lanczos steps and sampling over  $N_0 = 200$ – $300$  random states were sufficient to obtain reasonably accurate (within 5%) results for spin conductivity and static spin susceptibility.

### III. NUMERICAL RESULTS AND CONCLUSIONS

Using the finite-temperature Lanczos method we computed the spin-diffusion constant of the  $t$ - $J$  model in the physically relevant parameter regime, i.e., at  $J/t = 0.3$  and for four different hole dopings,  $\delta = 0, 1/16, 1/8$ , and  $1/4$ . The calculation was performed on a  $4 \times 4$  cluster with periodic boundary conditions.

In Fig. 1 we present numerical results for the renor-

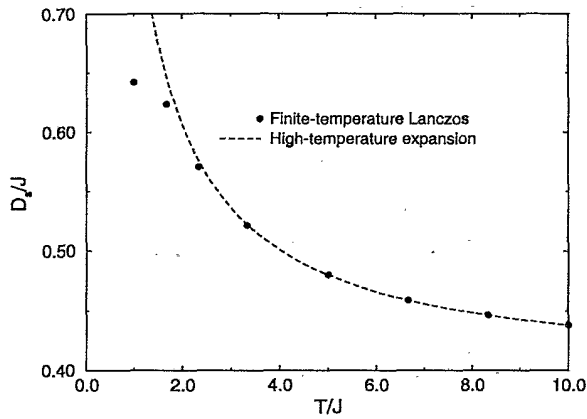


FIG. 1. Spin-diffusion constant  $D_s$  of the Heisenberg model vs temperature obtained by the finite-temperature Lanczos method (full circles). For comparison we present the high-temperature-expansion results obtained by Sokol *et al.* (Ref. 2) (dashed line).

malized spin-diffusion constant  $D_s/J$  of the Heisenberg model as a function of  $T/J$ . For comparison we present on the same graph  $D_s/J$  obtained by the high-temperature expansion by Sokol *et al.*,  $D_s/J = \sqrt{\frac{\pi}{20}}(1 + \frac{21}{20}\beta)$ , with  $\beta = J/T$ . Numerical results agree well with the high-temperature predictions down to temperatures  $T/J \sim 1.5$ , where  $D_s$  obtained by the numerical method saturates. In the  $T \rightarrow 0$  limit,  $D_s$  has to diverge, since the Heisenberg model at  $T = 0$  has finite, nonzero spin stiffness.<sup>10</sup> Saturation of  $D_s$  is therefore a finite-size effect. Nevertheless, the agreement with the high-temperature expansion at higher temperatures is quite encouraging. In contrast to the finite-size  $T = 0$  calculations, where results are strongly size dependent, the finite-temperature Lanczos method even on a small cluster provides results which are correct in the thermodynamic limit. We expect results to be size independent as long as the temperature is larger than the average discrete level spacing  $\epsilon$ , which is a consequence of the finite-size system. At finite doping, where the low-lying energy sector becomes more dense, we expect size-independent results down to much smaller temperatures.

We now turn to a doped case. In Fig. 2 we plot  $D_s/J$  as a function of temperature for four different values of doping at  $J/t = 0.3$ . As predicted by the infinite-temperature moment expansion,  $D_s/J$  is at high temperatures,  $T \gtrsim t$ , increasing approximately linearly with doping  $\delta$  for  $\delta \ll 1$ . In contrast to the undoped case where  $D_s$  as a consequence of finite-size effects saturates around  $T \sim J$ ,  $D_s$  at finite doping diverges with decreasing temperature (even in the finite cluster). This is an indication of the formation of a quantum coherent state, which has a finite spin stiffness at  $T = 0$ .  $D_s$  at small doping,  $\delta \leq 1/8$ , displays a monotonic  $1/T$ -like temperature dependence, similar to the undoped case. Somewhat unexpected is the nonmonotonic temperature dependence of  $D_s$  at moderate doping,  $\delta = 1/4$ . At high temperatures,  $D_s/J$  decreases with falling temper-

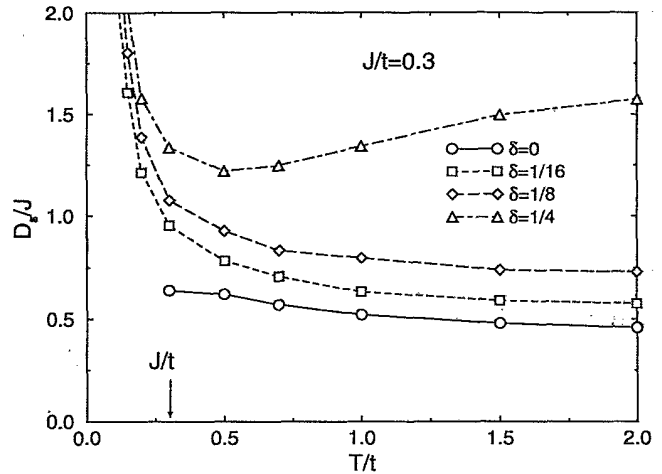


FIG. 2. Spin-diffusion constant  $D_s$  of the  $t$ - $J$  model vs  $T/t$  at  $J/t = 0.3$  and various doping values. The arrow above the temperature axis points to  $T = J$ . The accuracy of results at finite doping is 5%.

ature, reaches a minimum around  $T \sim 1.5 J$ , and when the quantum coherent regime sets in at even lower temperatures, sharply increases. We attribute this unusual nonmonotonic temperature dependence of the diffusion constant to the increase with doping of the number of low-energy short-lived many-body states. An enhanced density of low-energy states contributes to enhanced inelastic scattering, which consequently leads to a decrease of the diffusion constant. This picture is in agreement with the enhanced entropy at low temperatures<sup>11</sup> and strong charge-carrier scattering, which is manifested in the linear temperature dependence of the resistivity in the  $t$ - $J$  model.<sup>12</sup>

In conclusion, we have calculated the spin-diffusion constant  $D_s$  for the  $t$ - $J$  model. In the high-temperature limit,  $D_s$  can be approximately calculated via the moment expansion of the spin-conductivity function at  $t = 0$  or  $J = 0$  and for small doping  $\delta$ . In the Heisenberg limit, this method gives the correct result for  $D_s$ . At small doping,  $\delta \ll 1$ , and at  $t = 0$  (the dilute Heisenberg model),  $D_s/J$  decreases linearly with doping, indicating that the spin-current carriers are localized spins. At  $J = 0$  and  $\delta \ll 1$   $D_s/t$  is increasing linearly with  $\delta$  which is consistent with the fact that the spin-current carriers in this limit are mobile holes.

At finite temperatures, we used a recently developed Lanczos method to obtain  $D_s$  as a function of temperature and doping. We found good agreement of  $D_s$  in the Heisenberg limit with the high-temperature expansion result at higher temperatures. In agreement with our analytical predictions,  $D_s$  is increasing approximately linearly with doping at  $J/t \sim 0.3$  in the high-temperature and low-doping regime. Since the diffusion constant decreases with hole doping in a dilute Heisenberg model, i.e., at  $t = 0$ , we can conclude that hopping processes contribute substantially to spin diffusion at high temperatures. At moderate doping,  $\delta = 1/4$ , and high temperature,  $T \gtrsim J$ ,  $D_s$  decreases with decreasing temperature.

This anomalous behavior can be associated with the recently reported anomalous spin dynamics of the doped  $t$ - $J$  model at low frequencies<sup>13</sup> and enhanced entropy at low temperatures.<sup>11</sup>

After submitting this work we have been made aware of an excellent paper, written by Kopietz,<sup>14</sup> in which he derives relations between the spin diffusion coefficient and spin conductivity.

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