f-sum rule for the spin conductivity in itinerant magnets

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We derive a rigorous *f*-sum rule for the spin conductivity $D(\omega)$, valid in the paramagnetic regime of any itinerant system with nearest-neighbor hopping, assuming only that the potential energy conserves the total spin at each site. Two such itinerant systems are the double-exchange and Hubbard models. According to the *f*-sum rule, the integral over $D(\omega)$ is proportional to the average, electronic kinetic energy and is inversely proportional to the static susceptibility. The elastic value $D(0)$ can be obtained directly from the width of the quasielastic peak in neutron-scattering measurements. For infinite dimensions dynamical mean-field theory implies that $D(\omega)$ is proportional to the optical conductivity $\sigma(\omega)$ and that implies the *f*-sum rule for $D(\omega)$ reduces to the well-known optical sum rule for $\sigma(\omega)$. Our results place severe constraints on the appropriate model to describe the magnetoresistive manganites. © *2002 American Institute of Physics.*
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Because most models in condensed-matter physics lack an exact solution, rigorous sum rules can help us to understand physical behavior and to estimate the error incurred by various approximations. One of the most useful sum rules is the *f*-sum rule for the optical conductivity $\sigma(\omega)$. For a tight-binding model with nearest-neighbor hopping, this sum rule relates the integral over $\sigma(\omega)$ to the average kinetic energy $\langle K\rangle$ of the electrons. A correlated, nondisordered electronic system at zero temperature is an insulator if $\lim_{\omega\to 0}\ \sigma(\omega)=0$.[1]

This paper provides a generalization of these ideas to the paramagnetic spin conductivity $D(\omega)$ which was first introduced by Maleev[2] for a Heisenberg ferromagnet above T_C. Assuming nearest-neighbor hopping and that the potential energy V does not change the total spin at any site, the integral over $D(\omega)$ is shown to be proportional to $\langle K\rangle$ and inversely proportional to the static susceptibility χ. The elastic spin conductivity or spin-diffusion coefficient (SDC) $D_s=D(0)/2$ can be obtained directly from the width Γ of the quasielastic-peak in neutron-scattering measurements around $\omega=0$. In the hydrodynamic limit,[3] $\Gamma(\mathbf{k})=2/\tau(\mathbf{k})=2D_sk^2$ provides the lifetime $\tau(\mathbf{k})$ of a magnetic fluctuation with wave vector $\mathbf{k}$. When D_s becomes very small, such as near a ferromagnetic transition, the system can be considered a spin insulator[4] even when the particle conductivity $\sigma(0)$ remains high.

Recent work[5,6] in infinite dimensions suggests that spin relaxation in itinerant systems is much more complicated than in Heisenberg systems. For infinite dimensions, we use the dynamical mean-field theory[7] to show that $D(\omega)$ is proportional to $\sigma(\omega)$ and that the *f*-sum rule for $D(\omega)$ reduces to the well-known optical sum rule for $\sigma(\omega)$.

Due to the isotropy of magnetic fluctuations above T_C, the total spin correlation function $S(\mathbf{R}_i-\mathbf{R}_j,t)=\langle S^z_{\mathrm{tot},i}(t)S^z_{\mathrm{tot},j}(0)\rangle$ of a paramagnet does not contain any off-diagonal matrix elements. Generally, the Fourier transformed correlation function may be written as

$$S(\mathbf{k},\omega)=\frac{2}{1-e^{-\beta\omega}}\chi_2(\mathbf{k},\omega), \tag{1}$$

$$\chi_2(\mathbf{k},\omega)=\frac{1}{2}\sum_i\int dt\, e^{i\omega t-i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\times\langle[S^z_{\mathrm{tot},i}(t),S^z_{\mathrm{tot},j}(0)]\rangle. \tag{2}$$

In the hydrodynamic limit of small momenta and low frequencies,[3,8] the response function $\chi_2(\mathbf{k},\omega)$ can be parametrized by the form

$$\chi_2(\mathbf{k},\omega)=\frac{\omega k^2D_s\chi}{\omega^2+(D_sk^2)^2}, \tag{3}$$

which implies that $S(\mathbf{k},\omega)$ contains a quasielastic peak with width $\Gamma(\mathbf{k})=2D_sk^2$.

In place of Eq. (3), we employ a somewhat more general relation[2,8] which is valid for small k but arbitrary ω:

$$\chi_2(\mathbf{k},\omega)=\mathrm{Im}\,\chi(\mathbf{k},\omega+i\eta)=\mathrm{Im}\left.\frac{ik^2\overline{D}(u)\chi}{u+ik^2\overline{D}(u)}\right|_{u=\omega+i\eta}, \tag{4}$$

$$\overline{D}(u)=\int\frac{d\omega}{2\pi i}\frac{D(\omega)}{\omega-u},\quad \mathrm{Im}\,u>0, \tag{5}$$

where $\eta>0$ is infinitesimal. This parametrization of $S(\mathbf{k},\omega)$ and $\chi_2(\mathbf{k},\omega)$ is valid for $ka\ll 1$ and $k\xi\ll 1$, where ξ is the magnetic correlation length and a is the lattice spacing. So for a fixed $k\ll 1/a$, it fails only in the critical regime very close to T_C, when ξ becomes large. Comparing Eqs. (2) and (4), we confirm that the SDC is given by $D_s=D(0)/2$.

It follows that the spin conductivity can be written in terms of the spin-current response function $\Pi(\omega)$ as

$$D(\omega)=\frac{2\omega}{\chi}\lim_{k\to 0}\frac{1}{k^2}\chi_2(\mathbf{k},\omega)=-\frac{2a^2}{\omega\chi}\mathrm{Im}\,\Pi(\omega), \tag{6}$$

$$\Pi(\omega)=-\frac{i}{N}\lim_{k\to 0}\int_0^\infty dt\, e^{i\omega t}\langle[J_\gamma^z(\mathbf{k},t),J_\gamma^z(-\mathbf{k},0)]\rangle, \quad (7)$$

where γ is fixed and arbitrary (owing to the isotropy above T_C) and J_γ^δ is the spin current in the direction γ with polarization δ. These relations imply that $D(\omega)\geqslant 0$ for all frequencies.[8]

Consider a tight-binding Hamiltonian with the form $H=K+V$, where the kinetic energy operator is

$$K=-t\sum_{\langle i,j\rangle}(c^\dagger_{\mathbf{R}_i\alpha}c_{\mathbf{R}_j\alpha}+c^\dagger_{\mathbf{R}_j\alpha}c_{\mathbf{R}_i\alpha})=\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}c^\dagger_{\mathbf{k},\alpha}c_{\mathbf{k},\alpha}. \quad (8)$$

Here $c^\dagger_{\mathbf{R}_i\alpha}$ and $c_{\mathbf{R}_i\alpha}$ are the creation and destruction operators for an electron with spin $\alpha=\uparrow$ or $\downarrow$ at site i. For simplicity, we have assumed a single band but the f-sum rule derived below still holds for multiple bands so long as the hopping remains between nearest neighbors. If the system contains both local and electronic spin degrees of freedom, then the total spin at site i can be written $\mathbf{S}_{\mathrm{tot},i}=\mathbf{s}_i+\mathbf{S}_{\mathrm{loc},i}$, where $\mathbf{s}_i=(1/2)c^\dagger_{\mathbf{R}_i\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{\mathbf{R}_i\beta}$ is the electronic spin. The potential energy V is assumed to conserve the total spin at site i so that $[V,\mathbf{S}_{\mathrm{tot},i}]=0$. Both the Hubbard and double-exchange models obey these assumptions.

The spin current is obtained from the equation of continuity for the total spin:

$$i\frac{dS^\delta_{\mathrm{tot}\,\mathbf{k}}}{dt}=[S^\delta_{\mathrm{tot}\,\mathbf{k}},H]=\mathbf{k}\cdot\mathbf{a}_\gamma J^\delta_\gamma(\mathbf{k}), \quad (9)$$

$$J^\delta_\gamma(\mathbf{k})=\frac{t}{2i}\sum_i e^{-i\mathbf{k}\cdot\mathbf{R}_i}c^\dagger_{\mathbf{R}_i\alpha}\sigma^\delta_{\alpha\beta}(c_{\mathbf{R}_i+\mathbf{a}_\gamma,\beta}-c_{\mathbf{R}_i-\mathbf{a}_\gamma,\beta})$$

$$=\frac{1}{2a}\sum_{\mathbf{k}'}v_{\mathbf{k}',\gamma}c^\dagger_{\mathbf{k}',\alpha}\sigma^\delta_{\alpha\beta}c_{\mathbf{k}'+\mathbf{k},\beta}, \quad (10)$$

where $\mathbf{a}_\gamma$ is one of $z/2$ primitive lattice vectors and $v_{\mathbf{k},\gamma}=\partial\epsilon_{\mathbf{k}}/\partial k_\gamma$ is the electron velocity. Based on the definitions of $\chi_2(\mathbf{k},\omega)$ in Eq. (2) and $J^z_\gamma(\mathbf{k})$ in Eq. (10), we find that

$$\int\frac{d\omega}{\pi}\omega\chi_2(\mathbf{k},\omega)=\frac{1}{N}\mathbf{k}\cdot\mathbf{a}_\gamma\langle[J^z_\gamma(\mathbf{k}),S^z_{\mathrm{tot},-\mathbf{k}}]\rangle, \quad (11)$$

but the commutator is given by

$$[J^z_\gamma(\mathbf{k}),S^z_{\mathrm{tot},-\mathbf{k}}]=\frac{t}{4}\mathbf{k}\cdot\mathbf{a}_\gamma\sum_i\{c^\dagger_{\mathbf{R}_i,\alpha}c_{\mathbf{R}_i+\mathbf{a}_\gamma,\alpha}+c^\dagger_{\mathbf{R}_i,\alpha}c_{\mathbf{R}_i-\mathbf{a}_\gamma,\alpha}\}, \quad (12)$$

so that

$$\frac{1}{\chi}\int\frac{d\omega}{\pi}\omega\chi_2(\mathbf{k},\omega)=k^2\int\frac{d\omega}{2\pi}D(\omega)=-\frac{(ka)^2}{2Nz\chi}\langle K\rangle, \quad (13)$$

where the first equality follows from Eq. (6).

Thus, we finally obtain the f-sum rule for the spin conductivity:

$$\int\frac{d\omega}{2\pi}D(\omega)=-\frac{a^2}{2Nz\chi}\langle K\rangle, \quad (14)$$

which relates the integral over $D(\omega)$ to the expectation value of the kinetic energy. Equation (14) is valid for all temperatures and in any dimension.

In infinite dimensions, the response function can be considerably simplified by using dynamical mean-field theory. Due to the absence of off-diagonal matrix elements above T_C and the lack of current vertex corrections[9] in infinite dimensions, the spin-current response function can be written as $\Pi(\omega)=\Pi(i\omega_m=\omega+i\eta)$ where[5]

$$\Pi(i\omega_m)=\frac{T}{2a^2N}\sum_{\mathbf{k},l}G(\mathbf{k},i\nu_l)G(\mathbf{k},i\nu_l+i\omega_m)v^2_{\mathbf{k},\gamma}. \quad (15)$$

Here $G(\mathbf{k},i\nu_l)_{\alpha\beta}=G(\mathbf{k},i\nu_l)\delta_{\alpha\beta}$ is the one-particle Green's function, which is a function of the Matsubara frequencies $\omega_m=2m\pi T$ and $\nu_l=(2l+1)\pi T$ at temperature T. It follows that $D(\omega)$ is simply related to the particle conductivity $\sigma(\omega)$ by $D(\omega)=\sigma(\omega)/2\chi e^2$ and that the SDC is related to $\sigma(0)$ by $D_s=\sigma(0)/4\chi e^2$. So in infinite dimensions, the f-sum rule for $D(\omega)$ is equivalent to the the optical sum rule for $\sigma(\omega)$:[10]

$$\int\frac{d\omega}{\pi}\sigma(\omega)=-\frac{2e^2a^2}{Nz}\langle K\rangle. \quad (16)$$

With the effective mass defined by $1/m^*=-(2a^2/z)\times\langle K\rangle/N_{\mathrm{el}}>0$,[10] the right-hand side reduces to the well-known expression $N_{\mathrm{el}}e^2/Nm^*=(\omega_p^2/4\pi)(V/N)$.

Now we briefly explore the consequences of the f-sum rule for the double-exchange model,[11,12] where the Hund's energy $V=-2J_H\Sigma_i\mathbf{s}_i\cdot\mathbf{S}_{\mathrm{loc},i}$ acts to align the local and itinerant spins. At least qualitatively, the double-exchange model is believed to describe the magnetoresistive manganites, which become metallic below T_C. Recently, the f-sum rule for the optical conductivity has been used to understand the physics of manganites below T_C and to appraise the limitations of the double-exchange model.[13–15] The f-sum rule for the spin conductivity provides similar insight into the appropriate models to describe the spin dynamics of the manganites above T_C.

Neutron-scattering measurements[16,17] on manganites like $La_xCa_{1-x}MnO_3$ with $x\approx 0.3$ indicate that D_s exhibits little temperature dependence as T approaches T_C. Moreover, spin diffusion seems to persist even below T_C, where the quasielastic peak in $\chi_2(\mathbf{k},\omega)$ coexists with spin-wave peaks. The f-sum rule for the spin conductivity can help determine whether such behavior can arise from an exact treatment of the double-exchange model in three dimensions. Assuming that the magnetic transition is second order, then χ diverges and the integral over $D(\omega)$ must vanish as $T\to T_C$. Since $D(\omega)\geqslant 0$ for all ω, this implies that $D(\omega)\to 0$ for every ω and, in particular, that $D_s\to 0$ 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 in the vicinity of T_C. This prediction cannot be reconciled with the measurements described above.

Therefore, it is necessary to go beyond the double-exchange model in order to explain the spin dynamics of the manganites even above T_C. It is now well-established[18] that

lattice and magnetic polarons play an essential role in the manganites. The inhomogeneities produced by polarons[19] may open other avenues of magnetic relaxation close to T_C. Nevertheless, it is difficult to understand how the divergence of χ at T_C can be reconciled with the persistence of spin diffusion close to T_C. Even if the sample phase separates into ferromagnetic and paramagnetic regions, the divergence of χ in the regions which become ferromagnetic should produce a dramatic change in the overall value of D_s.

To conclude, we have developed an *f*-sum rule for the spin conductivity which relates the integral over $D(\omega)$ to the average, electronic kinetic energy. This *f*-sum rule provides an important check on models for spin relaxation in itinerant systems.

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