

Theory of coexisting transverse-spin freezing and long-ranged antiferromagnetic order in lightly doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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We provide an explanation of the spin-freezing transition recently observed by Chou *et al.* [Phys. Rev. Lett. **71**, 2323 (1993)] in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ for $x \lesssim 0.02$. We propose that topological excitations of the two-dimensional Heisenberg quantum antiferromagnet having noncoplanar transverse components have a pair-interaction energy that qualitatively and quantitatively agrees with the observed values of spin-freezing temperature as a function of doping.

I. INTRODUCTION

One of the avenues that may aid in the understanding of the properties of high-temperature superconductors involves examining the magnetic correlations that exist in the CuO_2 planes when doped with carriers. Experiments may be employed to investigate the local magnetic character of these systems—some of the most useful are nuclear quadrupole resonance (NQR) studies where the nucleus provides a local probe^{1,2}—and from this information many features of the magnetic ordering and/or fluctuations may be identified. Further, through resonance frequencies, one can measure the coupling between the nucleus and the electric field gradient surrounding it, thus providing insight into the electronic charge distribution of the system.

The parent compound of the Bednorz-Müller high- T_c compound, viz., La_2CuO_4 , is an antiferromagnetic (AFM) insulator which undergoes a Néel-ordering transition at ~ 300 K. When the compound has trivalent La substituted for with divalent Sr for low doping levels, viz., less than 1% Sr, the AFM ordering persists with the Néel transition temperature (T_N) rapidly decreasing with increasing Sr concentration. Recent ^{139}La NQR experiments³ on lightly doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x \leq 0.02$) have suggested that an interesting *addition* to the AFM structure takes place. To be specific, they have measured data consistent with the coexistence of the AFM phase with long-ranged order *and* a spin-glass phase, and have noted that this latter state can be interpreted to be composed of frozen transverse (i.e., perpendicular to the direction of the staggered magnetization) spin components. The state may be unambiguously assigned to be that of a spin glass via the stretched exponential recovery of the nuclear magnetization.⁴ A schematic diagram of the low- x phase diagram is shown in Fig. 1, where a line separating the pure Néel state and the spin-glass coexisting state is denoted by T_f ; we shall refer to this line as the spin-freezing temperature. The focus of this paper is to (i) suggest the spin texture of this coexisting state,

and (ii) show that this spin texture predicts an onset of coexistence at T_f consistent with experiment.

To begin, let us summarize the experimental results. Chou *et al.*³ have measured the ^{139}La NQR for different dopings and temperatures in lightly doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ for low temperatures. They measured the ^{139}La NQR frequency spectrum, and the relaxation times for the system's magnetization to return to equilibrium. The splitting of the $2\nu_Q$ NQR peaks due to the internal field associated with the AFM ordering of the spins at the Néel temperature led to a doping-dependent ordering temperature $T_N(x)$ —this determination of $T_N(x)$ is in agreement with other measurements, such as (i) their observations of the spin-lattice relaxation rate, and (ii) that found using static susceptibility measurements.⁵ Further, a large enhancement of this splitting was found at very

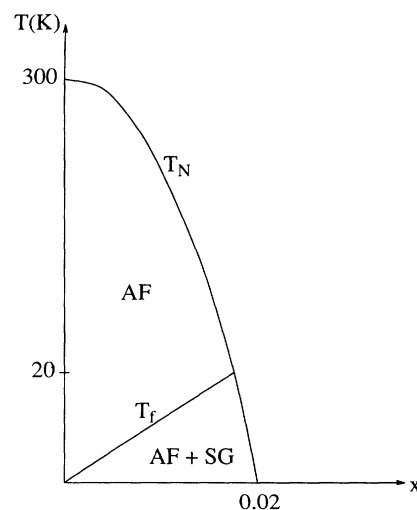


FIG. 1. Schematic phase diagram of the weakly doped system, showing that at temperatures below $T_f(x)$ a coexisting quantum AFM with long-ranged order and a spin-glass phase is observed.

low T , viz., at temperatures less than ~ 30 K, with the enhancement increasing with increasing x . Such behavior could be caused by an alteration of the underlying spin texture at this temperature. A change in the spin state may also be inferred from measurements of the relaxation rate of the $3\nu_Q$ transition—below ~ 70 K there is a large enhancement of this rate. The rate continues to increase until it rapidly drops off at very low temperatures, viz., temperatures around 5–16 K. Using this low-temperature behavior of the relaxation rate, Chou *et al.*³ identified the maximum of the NQR rate with the spin-freezing temperature T_f . As a function of doping it was determined to be approximately given by $T_f(x) \sim (815 \text{ K}) x$, and it is this linear relation that we have sketched in Fig. 1. Above 30 K (which may be interpreted to be the charge localization transition temperature⁶) the nuclear magnetization relaxed with a multiexponential decay, while below this temperature a dramatic change was noted in that now the relaxation of the magnetization was found to be described by $M(\infty) - M(t) \propto \exp[-(t/T_1^*)^{1/2}]$. This stretched exponential decay has been found in many systems studied by NQR (see, e.g., Ref. 4), and can be associated with a spin-glass ordering. However, the staggered moment of the AFM phase does not disappear, as indicated, e.g., in (i) the zero-temperature extrapolation of the $2\nu_Q$ splitting to the undoped value, or (ii) the continuous change of the splitting as T_f is traversed [see Fig. 1(c) of Ref. 3 for the data corresponding to both of these results], and thus the spin-glass ordering is found to coexist with the long-ranged AFM order.

Similar experimental results were found in both other NQR work⁷ at $x = 0.012$ and 0.015 , as well as in electron paramagnetic resonance (EPR) studies of the same system at $x = 0.009$ by Rettori *et al.*⁸ (see their Fig. 12)—this latter study again found an increase in internal field observed in the $-7/2 \rightarrow -5/2$ transition.

Clearly, it would be desirable to explain the above-mentioned behavior of the spin texture, and associated experimental measurements, and that is the focus of this paper. However, we wish to stress that this phenomenon is certainly not limited to the weakly doped Bednorz-Müller high- T_c compound or to AFM's. For example, previous experimental work on metallic spin glasses,⁹ and related computational studies¹⁰ on the moderately frustrated three-dimensional Heisenberg model, aids in the understanding of the high- T_c system we are studying. The work on the moderately frustrated Heisenberg model leads to the phase diagram shown in Fig. 2 of Ref. 10, and for dopant levels less than 25% is similar to that of our Fig. 1. In particular, the linear dependence of T_{xy} is the same. The authors of Ref. 10 identified the coexisting freezing of the transverse-spin degrees of freedom (transverse in the sense of being perpendicular to the already present ferromagnetic moment) and long-ranged ferromagnetic order at the spin-freezing temperature T_{xy} ; this latter ordering is not associated with a phase transition, only involving a short-ranged freezing of spins, and does not eliminate the ferromagnetic ordering. Unfortunately, these authors did not attempt to come to an analytic understanding of this similar linear dependence of the spin-freezing temperature on doping.

Nor did they attempt to justify the magnitude of the slope of this pseudophase boundary. That is precisely what we will present in this paper, albeit for a doped two-dimensional AFM.

Our paper is organized as follows. In Sec. II we summarize our previous work^{11,12} on the spin texture of singly and weakly doped CuO_2 planes; this involves studies of a noncoplanar spin texture where each Sr^{2+} dopant produces a spin state reminiscent of a singly charged Skyrmion. This section also serves to introduce the effective Hamiltonian that we will use to describe these doped planes. In Sec. III we present an analytic calculation of the two-Skyrmion interaction energy, and then use this to suggest the dependence of $T_f(x)$ on doping x . In Sec. IV we describe numerical work examining the same two-skyrmion interaction energy problem, and which reaches the same qualitative conclusion as in Sec. III, viz., an interaction energy $\propto x$, as well as providing a quantitative estimate of the slope. Then, we conclude this section by extrapolating the above work to determine the dependence of the spin-freezing temperature on doping via both a comparison to Chou *et al.*,³ and a simple counting of the degrees of freedom of the two-defect system. Finally, in Sec. V we summarize how this and other work on the weakly doped planes lead to a clear description of the novel spin texture of this system.

II. SPIN TEXTURE OF WEAKLY DOPED PLANES AT LOW T

Our model of the doped state of a CuO_2 plane at low T is based on a collection of partially delocalized holes. It is assumed that at low temperatures the carriers (viz., O holes) are not mobile, but rather are confined to some small region of a plane—this is entirely consistent with the localization transition seen in resistivity measurements between 50 and 100 K,¹³ as well as with the localization transition inferred from the NQR measurements of Chou.⁶ Physically, this localization can be considered to arise from the electrostatic attraction of the holes to a divalent Sr^{2+} ion.

A study of the magnetic spin texture of the ground state of the system for one hole was previously discussed by one of us,¹¹ within the framework of the t - J model. There, the hole was only free to move around one square plaquette, and it was found that the distorted AFM spins' state could be described, in a semiclassical sense,¹⁴ via a noncoplanar spin state with a rotational twist generated by the motional current of the hole circulating around the plaquette. Since the hole can circulate in either the clockwise or counterclockwise directions, the ground state is twofold degenerate. The combination of the above results allows one to infer that the spin texture is analogous to a singly charged Skyrmion found in classical chiral field theory.¹⁵ A numerical study¹⁶ found similar results in that the symmetry of the ground state in their work is identical to that of the Skyrmion.

In order to model many such partially localized carriers an effective Hamiltonian based on only the spin degrees of freedom was recently proposed by two of us.¹² Again,

based on the semiclassical representation of the spins, a Hamiltonian thought to mimic the effect of a hole confined to a single plaquette was used:

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{D}{S^4} \{ (\mathbf{S}_1 \cdot \mathbf{S}_2 \times \mathbf{S}_3)^2 + (\mathbf{S}_2 \cdot \mathbf{S}_3 \times \mathbf{S}_4)^2 + (\mathbf{S}_3 \cdot \mathbf{S}_4 \times \mathbf{S}_1)^2 + (\mathbf{S}_4 \cdot \mathbf{S}_1 \times \mathbf{S}_2)^2 \}. \quad (2.1)$$

The first term is the usual Heisenberg interaction representing the AFM background. J is the exchange coupling and $\langle ij \rangle$ denotes nearest neighbors in the plane; all near-neighbor pairs are summed over. The second term is introduced to generate a spin distortion that is the same as that produced by a hole circulating around the plaquette, the corners of the plaquette being denoted by 1, 2, 3, and 4. It was motivated by the work of Wen, Wilczek, and Zee¹⁷ on chiral spin liquids, and we shall refer to this term as the chiral-plaquette interaction. The prefactor of the interaction, D/S^4 , includes the $1/S^4$ factor to ensure that the Heisenberg and one-plaquette interactions both scale as S^2 . In Ref. 12 it was found that the interaction strength D must exceed $\sim 2.2J$ to be able to induce a singly charged Skyrmion ground state.

In order to model a plane doped with many O holes, one simply sums the chiral-plaquette interactions over all plaquettes having a Sr^{2+} ion above them; we distribute these defects randomly in the plane, and the resulting system is necessarily spatially inhomogeneous. The results of Monte Carlo studies of the spin-spin correlation function¹⁸ for large lattices at temperatures greater than $J/2$ were consistent with the empirical relation

$$\xi^{-1}(x, T) = \xi^{-1}(x, 0) + \xi^{-1}(0, T) \quad (2.2)$$

for the spin correlation length $\xi(x, T)$ found experimentally by Keimer *et al.*¹⁹ Note that in order for Eq. (2.2) to be obtained, it was necessary to use $D \sim 3J$. Further, in Ref. 12 we showed that a mean field model of this effective Hamiltonian was also consistent with Raman scattering experiments.²⁰ Now, in this paper, we extend our study of this effective Hamiltonian and demonstrate that the transverse spin-freezing temperature T_f may be quantitatively determined.

III. SEMICLASSICAL EVALUATION OF T_f

Here we present a discussion that leads to the analytic prediction for the dependence of the spin-freezing temperature on doping. The theory is based on a calculation of the interaction energy between two Skyrmions, and is performed utilizing the semiclassical approach of Shraiman and Siggia.¹⁴ Two of us²¹ have previously used a similar approach to calculate the interaction energy between two spin polarons generated in the frustrated bond model,^{22,23} a calculation that was also verified via exact diagonalization numerical work.²¹

To begin, we introduce the spin texture associated with one Skyrmion, where the spins are described by a contin-

uum field which is chiral. The direction of the staggered magnetization is denoted by a chiral vector field $\hat{\Omega}$ satisfying

$$\hat{\Omega} \cdot \hat{\Omega} = 1 \quad (3.1)$$

at every point in the plane. For a singly charged Skyrmion ($Q = \pm 1$) placed at the origin of the coordinate system in use, and for one choice of phase (see below), where the staggered magnetization is chosen to be in the positive z direction at infinity, the chiral field is found to be¹⁵

$$\hat{\Omega} = \frac{2\lambda x}{\lambda^2 + r^2} \hat{x} \pm \frac{2\lambda y}{\lambda^2 + r^2} \hat{y} + \frac{r^2 - \lambda^2}{r^2 + \lambda^2} \hat{z}, \quad (3.2)$$

where $r = \sqrt{x^2 + y^2}$ is the distance in the two-dimensional plane from the center of the Skyrmion, and λ is the size of the Skyrmion. That λ is the size of the Skyrmion is seen from the z component; viz., for $r < \lambda$ the direction of the staggered magnetization is reversed; formally, λ sets the length scale in the continuum problem. Further, upon changing the topological charge Q from 1 to -1 , the handedness of the twisting of the staggered magnetization is found to change. This is seen in the x and y components: Note that $\hat{\Omega}_x \pm i\hat{\Omega}_y \propto x \pm iy$, and thus the twisting of the staggered magnetization is described by a vector which circulates in the counter-clockwise ($Q = 1$) or clockwise ($Q = -1$) directions. Last, note that these two components of the spin field represent the transverse degrees of freedom and in this paper we shall suggest that it is these components that lock together, due to the mutual interactions between Skyrmions mediated by the Heisenberg AFM superexchange, leading to the spin freezing observed experimentally. A schematic representation of the spin texture for the $Q = 1$ Skyrmion is shown in Fig. 2.

We place one Skyrmion at $\mathbf{r} = 0$, and then a second at $\mathbf{r} = (\ell, 0)$. A charge of $Q = 1$ is chosen for the first, and the charge of the second will be determined by minimizing the interaction energy. The latter may be obtained from extrapolating the semiclassical theory¹⁴ to describe this system. There, in the classical approximation, Shraiman and Siggia derived that the interaction between a dipolar spin distortion \mathbf{P}_a , such as that which would be produced by a ferromagnetic bond in an AFM background (here $a = x$ or y labels a direction in the plane, such as the direction of the ferromagnetic bond), and the surrounding spins are given by

$$E_{\text{classical}} = -g_2 \sum_{a=x,y} \mathbf{P}_a \cdot (\hat{\Omega} \times \partial_a \hat{\Omega}). \quad (3.3)$$

The interaction strength g_2 is a phenomenological constant¹⁴ which in the $t \gg J$ limit is of $O(J)$. The ground state for the spin texture is then found by minimizing the sum of this energy and the usual Heisenberg AFM interactions between spins, the latter being most conveniently represented by the nonlinear σ model. We cannot associate a dipole moment with the spin texture of a Skyrmion, although it can be represented as a superposition of dipolar back-flow spin distortions induced by mobile holes subsequently localized onto a sin-

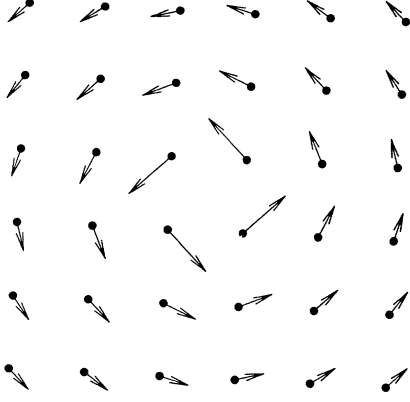


FIG. 2. Spin texture of a Skyrmion state, such as that given in Eq. (3.2), shown on a small 6×6 cluster (here, $Q=1$, and $\lambda = 0.5$). The spin state shown in the figure is found by taking Eq. (3.2) and rotating all spins (in spin space) by $\pi/2$ along the $+z$ direction. The spins at infinity are chosen to all point out of the page, and we have only displayed the projection of the spins onto the plane. Also, for ease of presentation, all spins on B sublattice sites of the background Néel state have been inverted ($\vec{S}_i \rightarrow -\vec{S}_i$), making the $D = 0$ state be a completely polarized ferromagnetic state. The solid dots denote the lattice sites.

gle plaquette.²⁴ A more direct way to represent the interaction of the background spin texture with the distortion induced by the Skyrmion is to replace

$$\mathbf{P}_a \rightarrow \hat{\Omega} \times \partial_a \hat{\Omega}, \quad (3.4)$$

where the staggered magnetization in this equation is that of the Skyrmion. This is the analog of the definition of the semiclassical dipole moment given by Shraiman and Siggia²⁵ for mobile holes. Then, we posit that one can represent the interaction between two Skyrmions as

$$E_{\text{int}} = -g_2 \sum_{a=x,y} \left(\hat{\Omega}^{(1)} \times \partial_a \hat{\Omega}^{(1)} \right) \cdot \left(\hat{\Omega}^{(2)} \times \partial_a \hat{\Omega}^{(2)} \right), \quad (3.5)$$

where the superscripts 1 and 2 denote each of the two Skyrmions. (Here we shall assume that each of the spin fields describing a Skyrmion is that produced by systems with only one defect. Thus, no back-reaction effects are included—these will be considered in the next section.) As an estimate of this energy we evaluate E_{int} at either of the points in space where either of the two Skyrmions are located. In the limit of large ℓ we obtain

$$E_{\text{int}} = -8J/\ell^2, \quad \ell \rightarrow \infty, \quad (3.6)$$

when the second Skyrmion is chosen to be a $Q = -1$ spin distortion, as this leads to the minimum energy. We note that the second Skyrmion can be placed *anywhere* in the lattice with the same result [viz., Eq. (3.6)] being obtained as long as the second Skyrmion has $Q = -1$. However, an arbitrary phase of the Skyrmion must be introduced [one that is not included in Eq. (3.2)]—see,

e.g., Ref. 15 for such a spin field], and then fixed via minimizing E_{int} with respect to this angle. (The interaction energy between two Skyrmions with the same topological charge scales as $1/\ell^4$ in the large- ℓ limit. As we will see below, this would lead to $T_f \propto x^2$, which indeed is not observed experimentally.)

For holes randomly placed on a two-dimensional lattice one expects that the doping x would be given by $x = 1/\ell^2$. This is consistent with the low-temperature neutron scattering studies of Keimer *et al.*¹⁹ where the correlation length $\xi(x, T)$ was found to behave approximately as $\xi(x, 0) \sim 1/\sqrt{x}$. Thus, expressing the two-Skyrmion interaction energy as a function of doping, we finally have

$$E_{\text{int}} = -8Jx. \quad (3.7)$$

Now, we suggest that one may associate the spin-freezing temperature T_f with this interaction energy; to be specific, $k_B T_f \propto |E_{\text{int}}|$. This is consistent with the notion that there is only short-ranged ordering of the transverse spins (similar to the results of Ref. 10) and the temperature at which the freezing occurs is such that at higher temperatures thermal fluctuations would destabilize a spin texture having these transverse degrees of freedom fixed. Of course, the energy associated with the state in which transverse degrees of freedom would become frozen into their ground state involves many Skyrmions, and thus we can only consider our treatment to be valid in the low doping limit. Noting that the spins are $S = 1/2$ quantum spins, and thus we must include the renormalization factor Z_c accounting for quantum fluctuations, we finally have that

$$k_B T_f \propto 8JZ_c x, \quad (3.8)$$

where $Z_c \sim 1.2$.²⁶ This relationship demonstrates that if the transverse degrees of freedom which freeze are associated with the many-Skyrmion spin texture produced by randomly distributed defects, a linear dependence of T_f on doping is expected. This is entirely consistent with the experiments of Chou *et al.*³

IV. NUMERICAL EVALUATION OF T_f

We have just seen that the freezing of the transverse degrees of freedom of two Skyrmions separated by a distance $1/\sqrt{x}$ would lead to a spin-freezing temperature linear in doping x . In this section we present a numerical procedure to obtain the quantitative dependence of T_f on the doping again in the low-temperature and lightly doped regime. This work is required since the relationship found in the previous section, viz., Eq. (3.8), did not allow for (i) the back reaction, and subsequent alteration, of a Skyrmion's core, and thus must be an overestimate of the interaction energy, and (ii) relies on an assumption for g_2 , the interaction strength. Here we wish to eliminate these limitations and determine a very accurate numerical relationship for T_f on x .

To establish a relationship between E_{int} and x we will consider a *lattice* of classical spins with two Skyrmions

placed somewhere in the lattice a distance ℓ lattice spacings apart. We calculate the interaction energy for such configurations by evaluating the ground state using Eq. (2.1) as the Hamiltonian (we have done the calculations for $S = 1$, and properly convert to $S = \frac{1}{2}$ below). In Ref. 12 it was found that the constant D was required to be $2.5 < D/J < 3.3$ in order to explain both the Raman scattering data²⁰ and the neutron scattering experiments consistent with Eq. (2.2). Here we perform the evaluation of E_{int} for $D/J = 2.5, 3$, and 3.3 in Eq. (2.1) to see how E_{int} varies with D/J .

The method employed in the evaluation of E_{int} is straightforward and involves studying $L \times L$ lattices with open boundary conditions with two chiral-plaquette interactions included on two plaquettes, and then extrapolating to $L \rightarrow \infty$; for concreteness, we explain the method by considering Fig. 3. In this figure we have shown a 6×6 lattice and have labeled two plaquettes S_1 and S_2 , where the separation between the centers of these plaquettes corresponds to $\ell = 2$. First, we find the single-skyrmion energy for finite L by eliminating the chiral-plaquette interaction on S_2 , and then find the ground state spin configuration and associated energy (with open boundary conditions)—denote this energy by $E_1(L, \ell)$, where the inclusion of ℓ in E_1 indicates that the Skyrmion is off center on the lattice. Then, the chiral-plaquette interaction on S_2 is reintroduced and the two-Skyrmion energy may be found once the two-Skyrmion ground state energy, denoted by $E_2(L, \ell)$, is known (again evaluated for open boundary conditions). All zero-force spin configurations were evaluated using a conjugate-force method similar to that employed in Ref. 21.

Consistent with the above semiclassical work, we always find that the ground state for two defects corresponds to opposite topological charges on the plaquettes. Then, in analogy to a two-particle binding energy, one defines

$$E_{\text{int}}(L, \ell) = E_2(L, \ell) - 2[E_1(L, \ell) - E_0(L)] - E_0(L), \quad (4.1)$$

where $E_0(L)$ is the energy of a pure Heisenberg AFM, with open boundary conditions, for an $L \times L$ lattice.

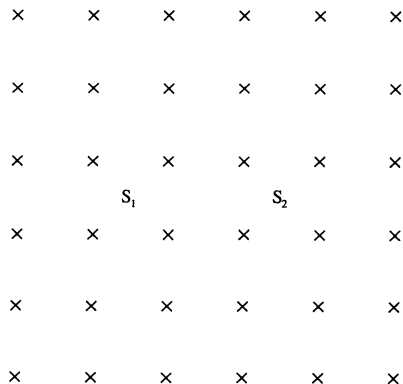


FIG. 3. Schematic diagram of a small cluster denoting two Sr defects above plaquettes S_1 and S_2 .

Finally, the large- L limit is taken to evaluate the interaction energy. An example of this extrapolation is shown in Fig. 4 for $D/J = 3$ and $\ell = 8\sqrt{2}$, the latter corresponding to $x = 0.0078$.

In Fig. 5 we have plotted $|E_{\text{int}}|/J$ vs x , utilizing $\ell = 1/\sqrt{x}$, for all three ratios of D/J . We obtain $5.14x \pm 0.08$, $5.00x \pm 0.08$, and $4.74x \pm 0.08$, for $D/J = 3.3, 3.0$, and 2.5 , respectively. The linear relation found in Eq. (3.8) is also found in the numerical work, and thus validates the hydrodynamic description of the spin degrees of freedom employed in the previous section. Further, it is clear that Eq. (3.7) represents the $D \rightarrow \infty$ limit (viz., no alteration of the Skyrmions' cores), and thus the energies found in this section are appropriately less than that of Eq. (3.7). It is encouraging that these energies do not change that much with D/J .

To determine the quantitative behaviour of $T_f(x)$ we must (i) transform the above energies to represent those of $S = \frac{1}{2}$ quantum spins, and (ii) determine the ratio of $|E_{\text{int}}|$ to $k_B T_f$. The first point may be incorporated by multiplying the interaction energies by $S(S+1)Z_c$. The second is a nontrivial exercise, and to resolve it one can appeal to experiment. Chou *et al.*³ investigated how best to describe the low-temperature maxima they observed in the NQR rate, and found that if one attempted to describe the maxima by a nonequilibrium freezing of the spins, with a correlation time τ associated with an Arrhenius law, viz.,

$$\tau \propto \exp(E/k_B T), \quad (4.2)$$

then the relaxation rate peaked at T_f with a related exponent

$$(E/k_B T_f) = 8.9 \pm 0.5 \quad (4.3)$$

(see Fig. 3 of Ref. 3). Clearly, the energy E in the exponent in Eq. (4.3) must be associated with an interaction energy, and thus we incorporate $|E_{\text{int}}|$ into this equation. A theoretical explanation for this relationship follows from application of the equipartition theo-

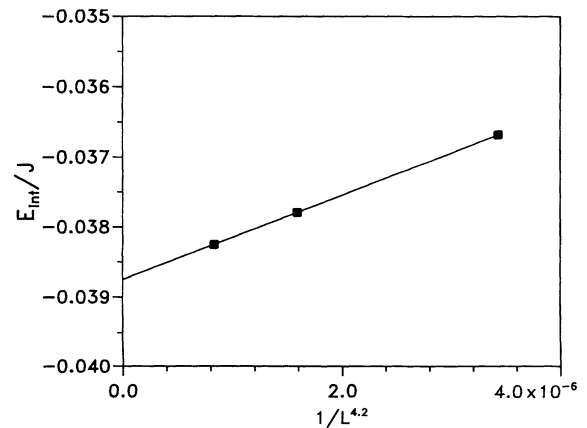


FIG. 4. Interaction energy defined in Eq. (4.1), in units of J , for $D = 3$ and a separation of defects of $8\sqrt{2}$ (viz., eight diagonal spacings), as a function of system size L for $L = 20, 24$, and 28 , showing the extrapolation to the bulk limit.

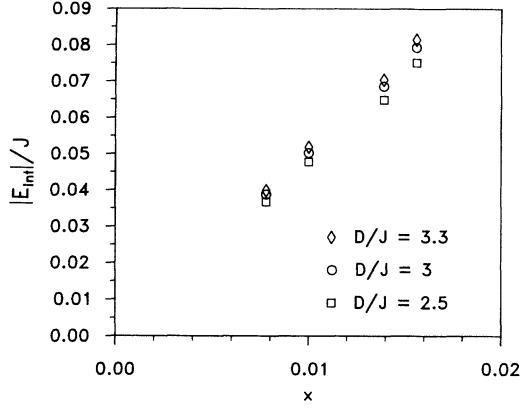


FIG. 5. The magnitude of the interaction energy $|E_{\text{int}}|$, in units of J , as a function of doping x for differing ratios of D/J . For each D/J , the data extrapolate linearly back to the origin.

rem. Apart from continuous symmetries, there are three differing low-energy states of the single defect problem: $Q = \pm 1$, as well as an excitation with zero topological charge (see Ref. 11 for a discussion of this important low-energy excitation). Thus, there are 3^2 static degrees of freedom for the two-defect system. However, since the spins are quantum spins, one should include a kinetic or dynamical component for the quantum fluctuations associated with each of these degrees of freedom. Then, counting $\frac{1}{2}k_B T$ for each such degree of freedom one expects

$$E_{\text{int}} = 2 \cdot 3^2 \cdot \frac{1}{2} k_B T_f. \quad (4.4)$$

This is consistent with the experimental determination of Chou *et al.* given in Eq. (4.3).

Then, we immediately have estimates for the spin-freezing temperature:

$$T_f \sim (805 \text{ K})x \quad \text{for } D = 3.3, \quad (4.5)$$

$$T_f \sim (784 \text{ K})x \quad \text{for } D = 3.0, \quad (4.6)$$

$$T_f \sim (743 \text{ K})x \quad \text{for } D = 2.5, \quad (4.7)$$

where we have used $J \sim 1550 \text{ K}$ for the exchange constant. We anticipate error bars of at least 5% on the above values, the major uncertainties coming from Z_c , and the exchange constant J .

The experimental result, viz., $T_f \sim (815 \text{ K})x$, shows that for D being close to 3 this two-Skyrmion interaction model provides a very credible estimate of the spin-freezing temperature. We stress that the temperature and doping dependence of the spin correlation length was found to agree with Ref. 19, and the Raman scattering spectra of Ref. 20, when the interaction strength $D \sim 3$ was used. It is gratifying that quantitative aspects of all three experiments seem to be consistent with the spin

texture found when the low-temperature doped planes are described by Eq. (2.1).

V. DISCUSSION

We have considered a simple model of the ground state spin texture produced by a Sr^{2+} defect and a single hole localized in its region,¹¹ and then utilized a model that allows for the description of many such defects,¹² viz., Eq. (2.1), where the chiral-plaquette interaction is summed over all plaquettes having a divalent Sr above them. We have used a semiclassical theory, and a numerical simulation technique, to determine the interaction energy between two such Skyrmion states. We find that the ground state of such a system corresponds to two Skyrmions having opposite topological charges, and that then the interaction energy, analogous to a two-particle binding energy, is proportional to $1/\ell^2$, as in Eq. (3.6), where ℓ is the separation between defects. Then, we associate the doping x with this average distance via $\ell = 1/\sqrt{x}$, consistent with the low-temperature spin correlation length determined by neutron scattering studies. The interaction energy is then found to scale linearly with doping, and if we associate the spin-freezing temperature with this interaction energy we can reproduce the same linear dependence that is observed experimentally. We have quantified this relationship by using the experimental value for the Arrhenius exponent given in Eq. (4.3). We can also present theoretical arguments that lead to values for the ratio of E to $k_B T_f$ that give the same numerical value as in Eq. (4.3) (e.g., a simple equipartition theorem argument also gives a ratio of 9).

Alternatively, the frustrated bond model of Aharony *et al.*²² predicts an interaction energy which scales linearly as x (see Ref. 21), but the spin texture of the ground state depends on the location of the frustrated bonds in the plane (e.g., see Ref. 21). Also, the ratio of Eq. (4.4) would be 4 for the frustrated bond model, and not ~ 9 , as found experimentally. However, it is not clear whether or not the experimental results of Chou *et al.*³ depend on probe frequency, and thus it is not clear if this particular kind of experiment could distinguish between these two models of the spin texture of the weakly doped planes. Since the important physics of both these models is that defects produce long-ranged spin distortions, and there is some evidence that such distortions are also produced by mobile holes at moderate doping levels,^{12,27} it is encouraging that both models rely on spin deviations coupling the perturbing effects of the holes to the long-wavelength spin waves, thus further justifying our hydrodynamic approach.

We have shown that for our model, with the interaction strength $D/J \sim 3$ in Eq. (2.1), the dependence of T_f on x is reproduced quite accurately. Further, since our model with this same ratio of D/J also reproduces the neutron scattering measurements of the spin correlation length^{19,28} and the Raman scattering spectra,²⁰ it seems likely that these novel topological excitations of the $S = \frac{1}{2}$ quantum AFM do indeed play a role in forming the spin textures of the low-temperature, inhomogeneously doped CuO_2 planes.

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