Spin Diffusion in $RbMnF_3$ [†]

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The spin-diffusion coefficient has been measured in RbMnF₃ between room temperature $(3.55T_N)$ and the critical region $(1.028T_N)$. The relationship $D(\kappa a)^{1/2} = 22.47 \pm 0.73$ meV Å² is well obeyed over this temperature range, and at room temperature the diffusion coefficient has the value 12.86 ± 0.21 meV Å². These numerical results are compared with the predictions of dynamic scaling and with theoretical calculations based on the Heisenberg model.

INTRODUCTION

In the paramagnetic region, spin fluctuations which vary slowly in space and time, so that local thermodynamic equilibrium is maintained, undergo relaxation by means of a diffusion process.¹ For a general disturbance of the spin system, the diffusion model can be used to describe the decay of correlations between spins separated by large distances and observed at widely different times. The behavior of this part of the correlation function is determined by those components of its Fourier transform having wavelengths long compared to the "characteristic" range of correlations, and energies small compared to interatomic interaction energies. These components can be selectively studied in an inelastic neutron scattering experiment and analyzed to obtain the diffusion coefficient.

Spin diffusion in crystal lattices was first discussed by Bloembergen² in connection with the problem of nuclear-spin relaxation in solids. The inelastic scattering of neutrons arising from the diffusion of atomic spins was treated by Van Hove³ and numerical estimates of diffusion coefficients at high temperatures were first obtained, within the framework of the Van Hove theory, by de Gennes.⁴ Detailed calculations, appropriate to high temperatures, have been carried out by Mori and Kawasaki, ⁵ Bennett and Martin, ⁶ Tahir-Kheli and McFadden, ⁷ Blume and Hubbard, ⁸ and Morita.⁹ In the critical region, in addition to the general predictions of the dynamic scaling theory of Halperin and Hohenberg, ¹⁰ there are also available calculations by Kawasaki,¹¹ Résibois and Piette,¹² Krueger and Huber, ¹³ and Hubbard.¹⁴ The intermediate temperature region has been treated by Hubbard¹⁴ for the Heisenberg ferromagnet and calculations for the Heisenberg antiferromagnet have been provided by Reiter.¹⁵ In addition to the purely theoretical calculations, the diffusion process has been studied using computer-simulation methods by Windsor¹⁶; Evans and Windsor¹⁷; and Watson, Blume, and Vineyard.¹⁸

The diffusion coefficient has been measured at room temperature for MnF₂ by Cribier and Jacrot¹⁹ and for RbMnF₃ by Windsor, Briggs, and Kestigian.²⁰ For both these compounds, the measurement temperature corresponds to about $3.5T_N$ and thus the results refer to high-temperature behavior. Limited results for the diffusion coefficient in the intermediate temperature region have been obtained by Riste²¹ for Fe_3O_4 ($T_N < T < 1.2T_N$) and by Deniz, Paranjpe, and Goyal²² for KMnF₃ (1.5 T_N $\langle T \langle 2T_N \rangle$. In the critical region, measurements have been reported by Als-Nielsen, Dietrich, Marshall, and Lindgård²³ and by Dietrich and Als-Nielsen²⁴ for the case of terbium which, as these authors suggest, may be regarded in first approximation as a uniaxial ferromagnet. Iron has been studied extensively in the critical region by Jacrot, Konstantinovic, Parette, and Cribier²⁵; Passell, Blinowski, Brun, and Nielsen²⁶; and Collins, Minkiewicz, Nathans, Passell, and Shirane.²⁷ A recent analysis by Als-Nielsen²⁸ of the experiment of Collins et al.²⁷ suggests that these measurements were not made in the hydrodynamic region and thus that the reported values of the diffusion coefficient were in error. This difficulty has been overcome in the recent work of the Neutron Inelastic Scattering Group at Saclay and Tournarie²⁹ and in that of Kahn and Parette, ³⁰ who find excellent agreement with the calculations of Résibois and Piette.¹² The earliest measurements on nickel by Cribier, Jacrot, and Parette³¹ indicated appreciable inelasticity in the scattering near T_c . The recent work of Minkiewicz, Collins, Nathans, and Shirane, $^{\rm 32}$ as discussed by Minkiewicz, ³³ shows that while significant uncertainties exist in the temperature dependence of both the diffusion coefficient and the range parameter, the measurements are consistent with the relationship $D \propto \kappa^{1/2}$ (D is the diffusion coefficient and κ is the inverse range parameter) predicted by dynamic scaling.¹⁰

In this paper we present measurements of the spin-diffusion coefficient for the ideal Heisenberg antiferromagnet $RbMnF_3$ from room temperature down to the critical region.

1103

8

EXPERIMENTAL METHOD AND RESULTS

For a paramagnet in the hydrodynamic regime, the Fourier transformed spin-correlation function for the total magnetization is given by³⁴

$$C^{M}(\mathbf{\tilde{q}},\omega) = \int_{-\infty}^{\infty} dt \sum_{\mathbf{\tilde{R}}} e^{i\mathbf{\tilde{q}}\cdot\mathbf{\tilde{R}}-i\omega t} \left\langle S_{0}^{Z}S_{\mathbf{\tilde{R}}}^{Z}(t) \right\rangle$$
$$\propto \left[\hbar\omega/(1-e^{-\hbar\omega\theta}) \right] \chi\left(\mathbf{\tilde{q}}\right) \frac{Dq^{2}}{(Dq^{2})^{2}+(\hbar\omega)^{2}} \quad , \quad (1)$$

where \mathbf{q} and $\boldsymbol{\omega}$ are wave vector and frequency, respectively; $\mathbf{\tilde{S}}_0$ and $\mathbf{\tilde{S}}_{\mathbf{R}}$ are the spins at the origin and at position $\mathbf{\bar{R}}$; $\beta = 1/kt$; $\chi(\mathbf{\bar{q}})$ is the wave-vector-dependent susceptibility; and D is the conventional spin-diffusion coefficient.³⁵ In terms of this correlation function, the cross section for inelastic scattering of neutrons in which the neutron momentum loss is $\hbar \mathbf{\bar{q}} = \hbar \mathbf{\bar{k}}_i - \hbar \mathbf{\bar{k}}_f$ and the neutron energy loss is $\hbar \boldsymbol{\omega} = E_i - E_f$, is given by

$$\frac{d^2\sigma}{d\Omega dE_f} \propto \frac{k_f}{k_i} |f(\vec{\mathbf{q}})|^2 C^{M}(\vec{\mathbf{q}},\omega) \quad , \qquad (2)$$

where \vec{k}_i and \vec{k}_f are the neutron wave vectors before and after scattering and $f(\vec{q})$ is the magnetic form factor. The diffusion coefficient D can thus be obtained at a given value of \vec{q} from the halfwidth in energy of the cross section.

In practice, the net magnetic scattering intensity, corrected for the energy sensitivity of the detector system, is given by the convolution of the cross section with the experimental resolution function, and the diffusion coefficient is determined from a least-squares fit of the convoluted cross section to the observed intensity. A multiplicative instrumental constant is left free in the least-squares analysis and provides an internal self-consistency check by requiring that it remain unchanged as the temperature and \vec{q} are varied.

The wavelength-dependent susceptibility has been obtained from the spherical model, ³⁶ which, for the present case of *antiferromagnetic* exchange and for \vec{q} measured either from the origin or from a reciprocal-lattice point for the *chemical* cell, is given by

$$\chi\left(\mathbf{\vec{q}}\right) = \frac{g^2 \mu_B^2 / 2 J^{\text{WBK}}}{(\kappa a)^2 + v(\mathbf{\vec{q}}) - v(2\pi \mathbf{\vec{\tau}}_m)} \quad , \tag{3}$$

where J^{WBK} , the exchange constant in the notation of Windsor, Briggs, and Kestigian, ²⁰ has the experimental value 0.284 meV, ³⁷ $v(\vec{q})$ is the nearestneighbor Fourier transform

$$\sum_{n=1}^{6} e^{i\vec{q}\cdot\vec{p}_n} \quad (\vec{p}_n \text{ is the nearest-neighbor} \\ \text{lattice vector),} \quad (4)$$

 $2\pi \dot{\tau}_m$ is a reciprocal-lattice point of the antiferromagnetic cell, *a* is the chemical cell lattice parameter, and κ is the inverse range parameter characterizing *antiferromagnetic* short-range order. TABLE I. Inverse range parameter as a function of temperature.

T	T/T_N	$\kappa a = 0.476 \epsilon^{0.701}$	$\kappa a = g\mu_B / expt.^a$	$(2J\chi_s)^{1/2}$ calc. ^b
295	3.55	3.89	3.17	3.12
166.04	2.00	2.02	1.80	1.84
124.53	1.50	1.24	1.20	1.27
103.78	1.25	0.764		0.941
93.398	1.125	0.470		
85.322	1.028	0.163		
84.02	1.012	0.0909		
83.27	1.003	0.0344		

 ${}^{a}\kappa a$ computed from experimental value of χ_{s} .

 ${}^{b}ka$ computed from theoretical value of χ_s obtained by G. S. Rushbrooke and P. J. Wood, Mol. Phys. <u>6</u>, 409 (1963).

The q's appearing in Eqs. (1) and (3) are based upon the chemical cell; however, the transformation $\vec{q} = 2\pi \vec{\tau}_m + \vec{q}^*$ (shifting the origin to $2\pi \vec{\tau}_m$) shows that for small \vec{q}^* , Eq. (3) has the Ornstein-Zernicke form and thus that κ is indeed the "antiferromagnetic" inverse range parameter. Values of the inverse range parameter to be inserted in Eq. (3)were determined experimentally in the following way: Close to the Néel point, κ was obtained from direct measurement³⁸ of the static correlation function. At higher temperatures it was derived from the measured³⁹ value of the staggered susceptibility $\chi_s = \chi(2\pi \tau_m)$ using $(\kappa a)^2 = g^2 \mu_B^2 / 2J \chi_s$, which can be obtained from (3) by setting $\vec{q} = 2\pi \vec{\tau}_m$. Values given by both procedures are listed in Table I, where it can be seen that satisfactory agreement is obtained in the intermediate temperature region. In actual calculations, κa values derived from χ_s were used down to and including $T/T_N = 1.50$; below this the critical-region power-law expression³⁸ for κa was used.

At each temperature D was obtained for six different points in reciprocal space, of which three were in the vicinity of the origin along the [011] direction and three along the [100] direction near the (1, 0, 0) reciprocal-lattice point. The range covered was in most cases $0.10 \le q \le 0.27$ Å⁻¹. [For reference, note that the antiferromagnetic point $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ corresponds to $q = 1.288 \text{ Å}^{-1}$.] In order to satisfy hydrodynamic conditions (i.e., $q < \kappa$), the lowest temperature included in the present analysis was 93.4 °K, for which T/T_N is 1.125 and κ is 0.109. This is a borderline temperature; at higher temperatures, conditions are clearly hydrodynamic; below this point deviations increase at a rapid rate. Energy scans with $E_i = 48 \text{ meV}$ were made at fixed values of \vec{q} and covered a range for which the maximum extent was ± 3 meV, in steps of 0.25 meV. These limitations were im-

Турев	lą I	$D \pmod{\text{Å}^2}$	σ ^c
A	0.104	12,92	1.16
Α	0.149	12.76	0.46
Α	0.193	12.99	0.38
А	0.238	12.42	0.32
в	0.147	12.99	0.22
в	0.189	12.97	0.22
в	0.231	12.67	0.45
	Weighted av:	12.86 ± 0.22	1

TABLE II. Diffusion coefficient at 295 °K.^a

^aNumerical values of the diffusion coefficient include a factor of \hbar since the Lorentzian of Eq. (1) is written in terms of an energy variable instead of frequency. This follows the convention of Ref. 20. Values in units of cm²/ sec may be obtained by multiplying by 1.518×10^{-4} .

^bFor type A, the origin of \bar{q} is (1,0,0) and its direction is [100]; for type B, the origin of \bar{q} is (0,0,0) and its direction is [011].

 $^{\circ}$ Standard deviation from least-squares determination of D.

posed by the inability of the three-crystal method to measure high-energy transfers for small q near the origin and by the need to avoid phonon contamination in data collected near the (1, 0, 0) reciprocal lattice point.

Background corrections to the observed scattering were obtained by making duplicate runs at liquid-helium temperature, where the elastic magnetic scattering is concentrated at the Bragg positions and the inelastic scattering appears in the form of well-defined magnons. For points close to the origin of reciprocal space, the major contribution to the background comes from air scattering in the intense primary beam. Some of this scattering passes through the crystal and is attenuated by an amount that varies with temperature because of magnetic ordering, and some bypasses the sample and enters the detection system directly. The detection system was masked off so that only scattering originating in or passing through the sample was counted. This background (after subtracting a contribution from cosmic rays and instrumental noise) was multiplied by the ratio of the crystal transmission at the temperature of the experiment to the transmission at helium temperatures. At room temperature the background correction for points near the origin amounted to approximately 25-30% of the total scattering at $\omega = 0$, but decreased rapidly for $|\omega| > 0$; the apparent broadening in energy of the air scattering, which is elastic, is merely due to instrumental resolution. In the vicinity of the (1, 0, 0) reciprocal-lattice point, on the other hand, the background corrections are less than 10% at $\omega = 0$. As mentioned previously, contamination by single-phonon scattering is avoided by limiting the range of energy transfer. Multiphonon processes, however, would not be observed at low temperatures but might be expected to contribute at high temperatures. Examination of the background scattering at higher reciprocal-lattice points, where such scattering should be more intense, indicates that background from this source is probably negligible compared to the magnetic scattering near the (1, 0, 0). Further indication that background scattering has been satisfactorily accounted for is provided by the fact that integrated intensities measured in the two regions of reciprocal space and subject to very different sources of error, agree on an absolute basis after adjustment is made for the difference in magnetic form factor.

Room-temperature measurements of the diffusion coefficient as a function of q are presented in Table II. The variation of D with temperature is given in Table III. Values appearing in Table III are weighted averages of individual determinations at the various values of q, inasmuch as no significant trend with q was observed. The quoted errors are weighted rms deviations with weights determined by the standard deviations given by the leastsquares fits for individual q's.

DISCUSSION AND COMPARISON WITH THEORY

Room Temperature

The room-temperature value of the diffusion coefficient, 12.86 meV Å², is appreciably larger than the value 8.0±1.0 meV Å² previously reported by Windsor *et al.*,²⁰ but agrees well with estimates obtained from recent theoretical calculations. At high temperatures, an expression for the diffusion coefficient can be given in terms of the frequency moments of the correlation function or of the imaginary part of the wavelength-dependent susceptibility.⁴⁻⁶ In the notation of Bennett and Martin,⁶

$$D = \left(\frac{\pi}{2} \frac{1}{\chi} \frac{C_1^3(0)}{C_3(0)}\right)^{1/2} , \qquad (5)$$

where χ is the ordinary static susceptibility and the C_n are moments of the imaginary part of the susceptibility χ'' given by

TABLE III. Temperature dependence of the diffusion coefficient.

<i>T</i> (°K)	T/T_N	D (meV Å ²)	ка	$D(\kappa a)^{1/2}$
295.0	3.55	12.86 ± 0.21	3.164	22.88
166.0	2.0	16.61 ± 0.68	1.798	22.27
124.5	1.5	19.10 ± 0.56	1.198	20.90
103.8	1.25	26.06 ± 0.66	0.755	22.64
93.4	1.125	33.72 ± 1.10	0.464	22,97
		Wei	ghted av.	$= 22.47 \pm 0.73$

$$\hbar^{n}C_{n}^{2}(\vec{\mathbf{q}}) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \ (\hbar\omega)^{n} \chi''(\vec{\mathbf{q}},\omega) \quad . \tag{6}$$

In the infinite-temperature limit this expression for D has been shown by these authors and by Mori and Kawasaki⁵ to be given by

$$\hbar D = 0.66 [S(S+1)]^{1/2} J^{WBK} a^2 = 9.91 \text{ meV } \text{\AA}^2$$
(7)

This result has also been obtained by the self-consistent calculations of the high-temperature correlation function by Blume and Hubbard⁸ and agrees reasonably well with the computer-simulation calculations of Windsor.¹⁶

At room temperature the diffusion coefficient can be evaluated by means of Eq. (5) using the frequency moments recently obtained by Collins⁴⁰ from high-temperature series expansions. The result of this calculation for 295 °K is 13.25 meV Å².

A second estimate of the room-temperature value can be obtained from the calculation of Hubbard¹⁴ for the paramagnetic phase of the Heisenberg ferromagnet. At high temperatures the diffusion coefficient given by this calculation is linear with respect to the temperature variable $\theta = J/kT$ and thus can be represented by a law of the form $D = D_{\infty}(1$ $+a_1\theta + \cdots)$. This law, because of the high-temperature limit, is also applicable, with the same coefficient a_1 to the antiferromagnet, in which case J, and thus θ , are negative. For RbMnF₃, using the theoretical value for D_{∞} , one obtains a room-temperature value of D equal to 12.14 meVÅ².

Temperature Dependence

The behavior of RbMnF₃ in the critical region³⁸ has been found to be in accord with the dynamicscaling predictions of Halperin and Hohenberg.¹⁰ These authors point out that the characteristic frequency $\omega_{\kappa}^{M_{\chi}}(\mathbf{\tilde{q}})$ and the shape function $f^{M_{\chi}}$ for the *total* magnetization of the antiferromagnet coincide with those for the staggered magnetization in the hydrodynamic region below T_N . In the hydrodynamic region above T_N , they predict, therefore, that $\omega_{\kappa}^{M_{\chi}}(\mathbf{\tilde{q}}) = Dq^2 \propto \kappa^{3/2} f(q/\kappa)$ and thus that $Dq^2/\kappa^{3/2}$ must be a function only of q/κ and

$$D \propto \kappa^{-1/2} \quad . \tag{8}$$

This relationship is presumably valid close to T_N , where $\kappa \to 0$. Measurements in this region are difficult, however, inasmuch as q must be kept small in relation to κ to satisfy the hydrodynamic conditions and, at the same time, large enough to avoid overlap with nonmagnetic scattering produced by finite instrumental resolution.

In Fig. 1 we have plotted the diffusion coefficient as a function of $(\kappa a)^{-1/2}$ from room temperature to the critical region. The (extended) dynamic-scaling prediction of Eq. (8) is well obeyed over the whole range. The point at $T/T_N = 1.028$, which is not hydrodynamic, has been included in the graph inasmuch as the sensitivity of D to departures from ideal conditions is unknown. This value corresponds to data obtained for q/κ in the range 2.8-6.1, and for which the frequency distribution shows significant departure from Lorentzian shape. At still lower temperatures, q/κ rises quite rapidly for the experimentally accessible values of \tilde{q} and the shape function is markedly non-Lorentzian.

Résibois and Piette¹² and Joukoff-Piette and Résibois⁴¹ have calculated the zero-order homogeneous functions of κ/q that appear in the expressions for the characteristic frequency for both the staggered magnetization and the total magnetization of the Heisenberg antiferromagnet in the critical region. If $2\pi \tau_m$ is the reciprocal-lattice vector characterizing the staggered magnetization and if we denote by \bar{q}^* the quantity $\bar{q} - 2\pi \tau_m$, the two characteristic frequencies can be written

$$\omega_{\kappa}^{N}(\mathbf{\bar{q}^{*}}) = \omega_{0}^{N}(\mathbf{\bar{q}^{*}})f^{N}(\kappa/q^{*}), \qquad \mathbf{\bar{q}^{*}} \simeq 0 \qquad (9a)$$

$$\omega_{\kappa}^{M}(\vec{\mathbf{q}}) = \omega_{0}^{M}(\vec{\mathbf{q}}) f^{M}(\kappa/q) / f^{M}(\mathbf{0}), \qquad \vec{\mathbf{q}} \simeq 0$$
(9b)

where $\omega_0^N(\mathbf{\bar{q}}^*) = \beta(q^*)^{3/2}$, $\omega_0^M(\mathbf{\bar{q}}) = \alpha q^{3/2}$, and α and β are constants. Good agreement between theory and experiment for $q^* \simeq 0$ has already been noted in Ref. 38. In the hydrodynamic regime $\omega_{\kappa}^M(\mathbf{\bar{q}}) = Dq^2$ and Eq. (9b) can be written

$$D(\kappa a)^{1/2} = [a^{1/2} \alpha / f^{M}(0)] f^{M}(\kappa/q) (\kappa/q)^{1/2} , \qquad (10)$$



FIG. 1. Variation of the diffusion coefficient with temperature and inverse range parameter. Error bars represent $\pm 2\sigma$.

1106

TABLE IV. Theoretical calculation^a of temperature dependence of the diffusion coefficient and test of the constancy of the computed value of $D(\kappa a)^{1/2}$.

ка	T °K ^b	D	$D(\kappa a)^{1/2}$
	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	7.28	
3.164	295	9.24	16.44
2.412	210	10.22	15.90
1.695	160	12.04	15.68
1.061	118	15.70	16.17
0.640	100	21.70	17.36

^aReference 15.

^bEstimated from experimental temperature dependence of  $\kappa a$  in Table III.

For the range of  $\kappa/q$  covered by our experiments the computed⁴¹ value of the quantity  $f^{M}(\kappa/q)(\kappa/q)^{1/2}$ is essentially constant and equal to  $1.23 \pm 2\%$ . Thus, the theory predicts  $D(\kappa a)^{1/2}$  to be a constant in agreement with experiment. Inserting the experimental value of  $D(\kappa a)^{1/2}$  in Eq. (10) and using the computed value for  $[f^{M}(\kappa/q)/f^{M}(0)](\kappa/q)^{1/2}$ , one obtains a value of 20.9 meV Å^{3/2} for the constant  $\alpha$ . The constant  $\beta$ , obtained from characteristic frequency measurements at the Néel point is approximately⁴² 20 meV Å^{3/2}. The ratio  $\alpha/\beta$  obtained in this way is thus approximately unity, whereas the suggested⁴¹ theoretical value for the critical region is  $\simeq 0.4$ .

The correlation function of Eq. (1) can be written more generally in the form

$$C(\mathbf{q}, \omega) \propto [\hbar \omega / (1 - e^{-\hbar \omega \beta})] \chi(\mathbf{q}) F(\mathbf{q}, \omega) , \qquad (11)$$

where  $F(\mathbf{q}, \omega)$  is the shape function of the spectral density of the spin-relaxation function.³⁴ Using a kinetic theory approach, Reiter¹⁵ has calculated  $F(\mathbf{q}, \omega)$  for RbMnF₃ at a number of points in the Brillouin zone over the temperature range 1.25  $\leq T/T_N \leq 3.55$ . These shape functions, after convolution with our experimental resolution function, were found to agree closely with experiment.³⁹ The theoretical calculations of  $F(\mathbf{q}, \omega)$  were carried out using an approximate version of an iterative solution to the kinetic equations. Reiter¹⁵ has recently extended these calculations, in the same de-

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gree of approximation, to the diffusion coefficient. The computed value of  $D(\kappa a)^{1/2}$  has been found to be essentially constant over the range  $1.5 \le T/T_N \le 3.55$  as shown in Table IV. The absolute value is about 30% below the experimental value; however, this discrepancy is not unexpected in view of the approximations used.⁴³

From a purely phenomenological point of view, the data, from infinite temperature down to the critical region, are reasonably well represented by the simple formula

$$D(T) = D_{\infty} [\chi_{\alpha} / \chi(0)]^{1/4} , \qquad (12)$$

which, using Eq. (3) and the theoretical infinite temperature result  $D_{\infty} = 9.91 \text{ meV} \text{ Å}^2$ , becomes

$$D(T) = 9.91 \left\{ \left[ (\kappa a)^2 + 12 \right] / (\kappa a)^2 \right\}^{1/4} .$$
 (13)

The computed room-temperature value, for example, is found to be 12.1; approximately 6% lower than the experimental value of 12.86. At low temperatures, Eq. (13) can be expanded to give  $D(\kappa a)^{1/2} \simeq 18.4$ , which is to be compared with the experimental value of 22.5.

While no theoretical justification of (12) can be given, the expression for D can be obtained by means of the following "heuristic" argument: The diffusion coefficient measures the decay of longwavelength  $(q \sim 0)$  excitations. For an antiferromagnet we may expect that these fluctuations will decay into the inherently more stable modes with  $\dot{q} = 2\pi \tau_m$ . The decay rate, and hence D, will depend on the ratio  $\chi_s / \chi(0)$ , which measures the relative stability of the antiferromagnetic and ferromagnetic modes and thus we write quite generally

$$D(T) = (\text{const}) \times f[\chi_s / \chi(0)] \quad . \tag{14}$$

The form of the function f in (14) can be chosen by using the scaling prediction for the critical region  $D \propto \kappa^{-1/2}$  and the spherical model result  $\kappa^{-1/2} \propto \chi_s^{1/4}$ giving  $f[\chi_s/\chi(0)] = [\chi_s/\chi(0)]^{1/4}$ . The constant in (14) is set equal to  $D_{\infty}$  since  $\chi_s/\chi(0) \rightarrow 1$  at high temperature.

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