Exchange narrowing of NMR line shapes in randomly diluted magnetic systems

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An analysis of ^{19}F NMR linewidths in the randomly diluted magnetic system $KMn_xMg_{1-x}F_3$ is presented. It is shown that good agreement with measured linewidths can be obtained if in the usual asymptotic spin-diffusion assumption for the spin autocorrelation function $\langle S_{i_\alpha}(\tau)S_{i_\alpha}(0)\rangle_{av}\alpha\tau^{-d(x)/2}$, $d(x)$ is taken to be independent of x above the percolation concentration. Experimental results in the system $KNi_xMg_{1-x}F_3$ are also presented. These data exhibit striking differences with the behavior of isostructural $KMn_xMg_{1-x}F_3$ whose origin is discussed.

I. INTRODUCTION

Considerable effort has been dedicated to the understanding of the effect of dimensionality upon the exchange narrowing of resonance lines.[1,2] In dense paramagnetic systems the NMR line shape is often dominated by the combined effect of the hyperfine interaction and the strong exchange coupling between the electronic spins of the magnetic ions. The rapid exchange-induced modulation of the local magnetic field experienced by the nuclei has a narrowing effect upon the NMR line shape which becomes sensitive to the long-time asymptotic behavior of the electronic-spin autocorrelation function. Various arguments[3,4] of quite general nature strongly suggest that this asymptotic regime should be governed by spin diffusion. This would imply a strong dependence of the NMR linewidth upon the dimensionality of the magnetic system. In systems where the exchange coupling is predominantly along chains, for example, one expects exchange narrowing to be largely inhibited because of the important role of spin diffusion. A linewidth orders of magnitude larger than in tridimensional systems can be expected in this case, a prediction that appears to be borne out by some experimental observations.[1]

The spin-diffusion assumption leads to an electronic-spin auto correlation function at the ith site of asymptotic form

$$\langle S_{i_\alpha}(\tau)S_{i_\alpha}(0)\rangle \propto \tau^{-d/2} ,$$

where $\alpha = x$, y, or z, and d is the Cartesian dimensionality. Of considerable interest is the generalization of this concept to the fractal geometry of percolating clusters. The possibility of describing the asymptotic behavior of the spin autocorrelation function in a dilute Heisenberg magnet by what one may call an effective dimensionality $d(x)$ varying smoothly with x, was first examined by Klenin and Blume.[5] By means of computer simulations, these authors calculated the spin autocorrelation function at infinite temperature, for a Heisenberg magnet of classical spins. Quenched disorder was introduced by random substitution of a fraction $1-x$ of the magnetic ions by a nonmagnetic species. Although their calculations were limited to times shorter than $3/J$, where J is the nearest-neighbor exchange coupling, the results for $x=1$ correctly described the expected asymptotic behavior

$$\langle S_{i_\alpha}(\tau)S_{i_\alpha}(\alpha)\rangle \propto \tau^{-d(1)/2}$$

with $d(1)=3$. As x decreased, $d(x)$ appeared to decrease smoothly, but no quantitative statement could be made about a possible asymptotic behavior of form

$$\langle S_{i_\alpha}(\tau)S_{i_\alpha}(0)\rangle_{av} \propto \tau^{-d(x)/2}$$

for $x<1$.

The NMR measurements of Borsa and Jaccarino[6] in the randomly diluted magnetic system $KMn_xMg_{1-x}F_3$ are also quite revealing. In this cubic perovskite structure three ^{19}F NMR lines have been observed above the ordering temperature. They were assigned[6] to fluorine nuclei having both of their nearest neighbors magnetic (I_2), having only one magnetic nearest neighbor (I_1), or missing both magnetic nearest neighbors (I_0). The width of the I_0 resonance appears to be mainly determined by magnetic dipole-dipole interactions of ^{19}F nuclei with second-nearest neighbors and more remote electronic spins and also by dipolar interactions among nuclear spins.[6] In contrast the I_1 and I_2 resonances in $KMn_xMg_{1-x}F_3$ are predominantly broadened by a transferred hyperfine coupling and narrowed by the exchange interaction among Mn^{2+} ions. As a consequence, the width of these lines becomes considerably larger with increasing magnetic dilution reflecting an average reduction of the exchange frequency with decreasing x. In $KMn_xMg_{1-x}F_3$ the width of I_1 resonance is particularly interesting because it can be followed experimentally over a considerable range of concentrations.[6]

The possibility of employing these NMR data in the range $x_p<x<1$, where x_p denotes the percolation concentration, to test the conjecture of an effective dimensionality $d(x)$ varying smoothly with the concentration of magnetic ions, has been suggested by D'Ariano *et al.*[7] These authors found that an extrapolation of the results of Ref. 5 together with the assumption

$$\langle S_{i_\alpha}(\tau)S_{i_\alpha}(0)\rangle_{av} \propto \tau^{-d(x)/2}$$

for long times, could be reconciled with the experimental-

ly determined linewidth of the I_1 resonance in $KMn_xMg_{1-x}F_3$.

Our own analysis of these data which is presented in this paper does not support this interpretation. We conclude that if one assumes the asymptotic form proposed for the autocorrelation functions, the effective dimensionality defined above can be taken to be independent of x, at least for $x-x_p \geq 0.1$. We also report some new experimental NMR results in the randomly diluted magnet $KNi_xMg_{1-x}F_3$ which exhibit striking differences with the behavior of the isostructural compound $KMn_xMg_{1-x}F_3$. A comparison between both systems suggests that a percolation model quite different from the conventional site-dilution scheme, may be necessary to understand the behavior of $KNi_xMg_{1-x}F_3$.

II. EXPERIMENTAL RESULTS

Figure 1 shows a ^{19}F NMR spectrum in $KNi_xMg_{1-x}F_3$ with $x=0.65$ obtained at room temperature and at a frequency of 20 MHz. The sample was a single crystal and the external magnetic field was parallel to a [100] crystal axis. The ordering temperature for this particular sample as determined by NMR was 67 K. Using a conventional continuous wave NMR spectrometer with peak-to-peak field modulation amplitudes of up to 20 G and also using pulsed NMR, spectra were recorded for samples with $x=0.9$, 0.65, 0.35, 0.2, and 0.1 for various orientations of the crystals with respect to the external magnetic field. In

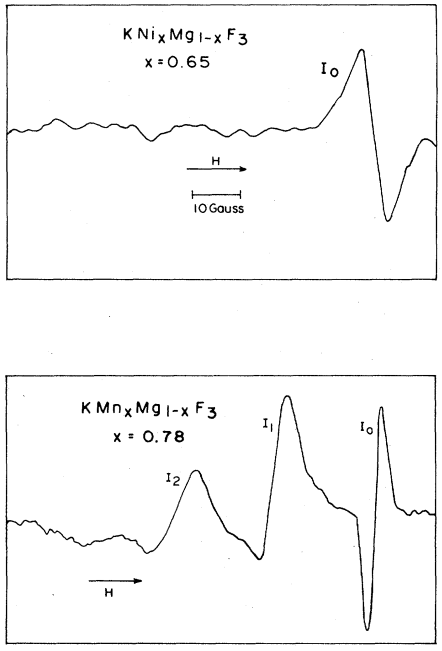

FIG. 1. (Top) ^{19}F derivative NMR spectrum in $KNi_xMg_{1-x}F_3$ with $x=0.65$ obtained at room temperature and at a frequency $\nu=20$ MHz. (Bottom) typical F^{19} NMR spectrum at room temperature in $KMn_xMg_{1-x}F_3$ with $x=0.78$ and $\nu=22$ MHz, from the work of D'Ariano and Borsa (Ref. 8). The horizontal scales in both spectra do not coincide.

all cases a single line was observed. From the concentration dependence of the amplitude of this resonance and from the negligible shift with respect to the ^{19}F Larmor frequency it was identified as the I_0 resonance. For comparison, Fig. 1 also shows a NMR spectrum in polycrystalline $KMn_xMg_{1-x}F_3$ for $x=0.75$ obtained by D'Ariano *et al.*[8] which clearly displays all three ^{19}F lines I_0, I_1, and I_2.

We conclude that in the concentration range of our experiments, the I_2 and I_1 resonances are much broader in $KNi_xMg_{1-x}F_3$ than in isostructural $KMn_xMg_{1-x}F_3$. Although I_1 and I_2 lines of somewhat larger widths may be expected in $KNi_xMg_{1-x}F_3$ at room temperature, because the ordering takes place at higher temperatures than in $KMn_xMg_{1-x}F_3$, we believe that this effect alone cannot explain the absence of these lines. This conclusion is supported by the behavior with temperature of the linewidth of the I_1 and especially the I_2 resonance in $KMn_xMg_{1-x}F_3$ which can be detected as close as 10–20 K from the ordering temperature.[8] Before attempting an explanation of our experimental results we present a line-shape analysis with the aim of accounting for the concentration dependence of the I_1 resonance line in $KMn_xMg_{1-x}F_3$.

III. LINE-SHAPE ANALYSIS

Exchange narrowing of NMR line shapes in paramagnetic systems can be conveniently treated within the framework of the Kubo-Tomita[9] theory. The relaxation function $\phi(\tau)$ whose Fourier transform represents the line shape is given by

$$\phi(\tau)=\exp\left[-\int_0^t (t-\tau)\psi(\tau)d\tau\right], \tag{1}$$

where the correlation function for fluctuations in the local magnetic field experienced by the nuclei denoted by $\psi(\tau)$, can be shown[1] to have the general form

$$\psi(\tau)=(1/N)\sum_{\mathbf{R}}\sum_{mm'} Q^{\delta}_{mm'}\Pi_{\mathbf{R}+\delta}(1-\Pi_{\mathbf{R}-\delta}) \times\langle S_{\mathbf{R}+\delta,m}(\tau)S_{\mathbf{R}+\delta,m'}(0)\rangle e^{im\omega_0}. \tag{2}$$

Given the cubic perovskite structure with lattice constant a, the position of a fluorine nucleus is labeled by the index $\mathbf{R}$ in Eq. (2). For every fluorine position $\mathbf{R}$, δ denotes a vector of length $a/2$ joining this position with that of a nearest-neighbor magnetic ion. Thus δ can point along any one of the three cubic axes. $\Pi_{\mathbf{R}+\delta}$ in Eq. (2) denotes the occupation number of the magnetic site at $\mathbf{R}+\delta$, i.e., $\Pi_{\mathbf{R}+\delta}=1$ if the site is occupied by a magnetic ion and $\Pi_{\mathbf{R}+\delta}=0$ otherwise. $Q^{\delta}_{mm'}$ is a quadratic expression in the components of the hyperfine tensor. For the fluorine sites selected by the factor $\Pi_{\mathbf{R}+\delta}(1-\Pi_{\mathbf{R}-\delta})$ which are those contributing to the I_1 resonance, $Q^{\delta}_{mm'}$ can be assumed to be independent of $\mathbf{R}$ and to depend only upon the orientation of the vector δ with respect to the external magnetic field. Moreover,

$$N=\sum_{\mathbf{R}}\Pi_{\mathbf{R}+\delta}(1-\Pi_{\mathbf{R}-\delta})$$

represents in Eq. (2) the total number of fluorine nuclei

contributing to the I_1 resonance. Other terms in Eq. (2) have the following meanings:

$$S_{\mathbf{R}+\delta,m}=S_{\mathbf{R}+\delta,x}\pm iS_{\mathbf{R}+\delta,y}$$

for $m=\pm 1$ and

$$S_{\mathbf{R}+\delta,m}=S_{\mathbf{R}+\delta,z}$$

for $m=0$; here $\mathbf{S}_{\mathbf{R}+\delta}(\tau)$ denotes the spin operator corresponding to the magnetic ion at site $\mathbf{R}+\delta$ whose time dependence is governed by an isotropic exchange interaction. The factor $e^{im\omega_0}$ may give rise to nonsecular broadening for $m\neq 0$ and originates in the noncommutativity between the hyperfine interaction and the electronic Zeeman energy. ω_0 denotes the electronic angular precession frequency in the external magnetic field assumed to be parallel to the z axis.

Since we are assuming for the fluorine nuclei contributing to the I_1 resonance a transferred hyperfine interaction with just a single magnetic nearest neighbor, only the autocorrelation functions

$$\langle \mathbf{S}_{\mathbf{R}+\delta,m}(\tau)\mathbf{S}_{\mathbf{R}+\delta,m'}(0)\rangle$$

are involved in Eq. (2). Correlations between electronic spins at different sites do not contribute to $\psi(\tau)$.

Since

$$\langle S_{\mathbf{R}+\delta,m}(\tau)S_{\mathbf{R}+\delta,m'}(0)\rangle$$

in Eq. (2) should be independent of the orientation of δ, whereas $Q^{\delta}_{mm'}$ only depends on δ and not on $\mathbf{R}$, one can substitute for $Q^{\delta}_{mm'}$ the quantity $\bar{Q}_{mm'}$ given by

$$\bar{Q}_{mm'}=(\tfrac{1}{3})(Q^{\delta_x}_{mm'}+Q^{\delta_y}_{mm'}+Q^{\delta_z}_{mm'}) .$$

Furthermore, we find it essential for our analysis to separate the sum over $\mathbf{R}$ in Eq. (2) into a sum of sorted[5] correlation functions

$$G_{mm'}(\tau,x)=(1/N)\sum_{\mathbf{R}}\langle S_{\mathbf{R}+\delta,m}(\tau)S_{\mathbf{R}+\delta,m'}(0)\rangle\Pi_{\mathbf{R}+\delta}(1-\Pi_{\mathbf{R}-\delta})$$
$$=(1/N)\sum_{l}\sum_{r=0}^{5}(6-r)\langle S^{(r)}_{lm}(\tau)S^{(r)}_{lm'}(0)\rangle\Pi^{(r)}_{l} . \tag{3}$$

In the right-hand side of Eq. (3) the sum runs over all magnetic sites in the simple-cubic lattice with lattice constant a. $\Pi^{(r)}_l$ is different from zero only if the site l is occupied by a magnetic ion having r nearest neighbors also occupied by magnetic ions. If $\Pi^{(r)}_l$ is different from zero, the factor $6-r$ with $r=0,1,\ldots,5$ counts the number of fluorine atoms hyperfine coupled to the magnetic ion at site l.

Equation (3) can be written in a more useful form in terms of the sorted autocorrelation functions averaged over the whole sample $\langle S^{(r)}_m(\tau)S^{(r)}_{m'}(0)\rangle_{\rm av}$. These are identical to those defined in Ref. 5. Noticing that the fractional number of fluorine atoms contributing to the I_1 resonance is

$$\mathcal{N}=\sum_{r=0}^{5}(6-r)6!x^r(1-x)^{6-r}/r!(6-r)!$$

one obtains from Eq. (3)

$$G_{mm'}(\tau x)=(1/\mathcal{N})\sum_{r=0}^{5}[6!(6-r)x^r(1-x)^{6-r}/r!(6-r)!]\langle S^{(r)}_m(\tau)S^{(r)}_{m'}(0)\rangle_{\rm av} . \tag{4}$$

The sorted autocorrelation functions $\langle S^{(r)}_\alpha(\tau)S^{(r)}_\alpha(0)\rangle_{\rm av}$ with $\alpha=x$, y, or z are known from the computer experiments of Klenin and Blume.[5] Their behavior is quite different for $0\leq\tau\leq 1/JS(S+1)$. At short times within this interval a much slower decay is observed for the smaller values of r. It is also observed that the sorted autocorrelation functions are quite independent of the actual concentration of magnetic ions. Furthermore, the decay of $\langle S^{(r)}_\alpha(\tau)S^{(r)}_\alpha(0)\rangle_{\rm av}$ in the time interval considered appears to become more independent of r at longer times. This is not unreasonable because in this asymptotic region the sorted correlation functions are expected to reflect the configuration of the cluster far away from the initial ion and therefore to become insensitive to the actual values of r.

We use the numerical values for the sorted autocorrelation functions given in Ref. 5 for the longest time available $\tau_0\sim 1.3/JS(S+1)$. Moreover, following the prescription of Gulley *et al.*[10] we write for $\tau>\tau_0$,

$$\langle S^{(r)}_m(\tau)S^{(r)}_{m'}(0)\rangle_{\rm av}=\langle S^{(r)}_m(\tau_0)S^{(r)}_{m'}(0)\rangle_{\rm av}(\tau/\tau_0)^{-d(x)/2} . \tag{5}$$

If $d(x)$ were actually independent of x and remained equal to $d(1)=3$, the only dependence of the linewidth upon concentration of magnetic ions would come from Eq. (4). This would yield the following expression for the linewidth $\delta H(x)$:

$$\delta H(x)=\frac{\delta H(1)\sum_{r=0}^{5}f^{(r)}(x)\dfrac{\langle S^{(r)}_\alpha(\tau_0)S^{(r)}_\alpha(0)\rangle_{\rm av}}{\frac{1}{3}S(S+1)}}{\sum_{r=0}^{5}f^{(r)}(x)} , \tag{6}$$

where

$$f^{(r)}(x)=6!x^r(1-x)^{6-r}/r!(6-r-1)! .$$

The values adopted for

$$\langle S^{(r)}_\alpha(\tau_0)S^{(r)}_\alpha(0)\rangle_{\rm av}/\tfrac{1}{3}S(S+1)$$

were 0.70, 0.42, 0.30, 0.23, and 0.17 for $r=1,2,\ldots,5$, respectively. Some of these values not explicitly given in Ref. 5 were obtained by interpolation.

Figure 2 shows the experimental results of Borsa and Jaccarino[6] in $KMn_xMg_{1-x}F_3$ for the linewidth of the I_1 resonance together with the theoretical prediction based upon Eqs. (4)—(6). It appears that the assumption $d(x)=d(1)=3$ leads to very good agreement with the experimental results, at least for $x-x_p \geq 0.1$.

The role of the sorted autocorrelation functions $\langle S_\alpha^{(0)}(\tau)S_\alpha^{(0)}(0)\rangle_{\rm av}$ needs some special clarification. If one assumes a nonzero exchange interaction only between magnetic nearest neighbors, this autocorrelation function would not decay. Its actual decay would be governed by weak exchange couplings with next-nearest or even more remote neighbors. Although most magnetic sites with $r\neq 0$ predominantly belong to the infinite cluster[11] for $x-x_p \geq 0.1$, the sites with $r=0$ are isolated and therefore are characterized by a completely different spin dynamics. Their role appears to be important in the spin-lattice relaxation process[12] of fluorine nuclei but not in the line shape, at least for the concentration range considered. The actual contribution of $\langle S_\alpha^{(0)}(\tau)S_\alpha^{(0)}(0)\rangle_{\rm av}$ is difficult to calculate accurately, but an estimate assuming maximum influence would increase the value of $\delta H(x)$ calculated through Eq. (6) by about 10% in the region $x-x_p \geq 0.1$.

It is worth pointing out that, unlike other calculations,[7] the one leading to the theoretical curve shown in Fig. 2 contains essentially no adjustable parameters or cutoffs to remove divergencies. The value $\delta H(1)=9.5$ G was not explicitly calculated but was chosen[6] as half the value of the linewidth in pure $KMnF_3$. Since this width can be accounted to a good approximation by the known values of the hyperfine coupling and exchange interaction using the same line-shape analysis outlined above,[10] the agreement can be considered quite satisfactory.

IV. DISCUSSION

From the results shown in Fig. 2 it is apparent that the variation in linewidth observed in $KMn_xMg_{1-x}F_3$ can be entirely accounted for by the concentration dependence of the weights given to the amplitudes of the sorted autocorrelation functions without any change in the effective dimensionality, at least in the concentration range $x-x_p \geq 0.1$. This conclusion is in contradiction with earlier suggestions.[7,8] In the critical region $0<x-x_p<0.1$, the existing data are not sufficiently reliable but may indicate a larger width than predicted by Eqs. (4)—(6). This may imply a smaller value of d in this region. If the conjecture of Alexander and Orbach[13,14] could be extended also to spin diffusion in a percolating cluster, one would actually expect the fraction dimensionality $d\approx\frac{4}{3}$ to substitute d in Eq. (5). This would lead to a much larger width at percolation than predicted by Eqs. (4)—(6).

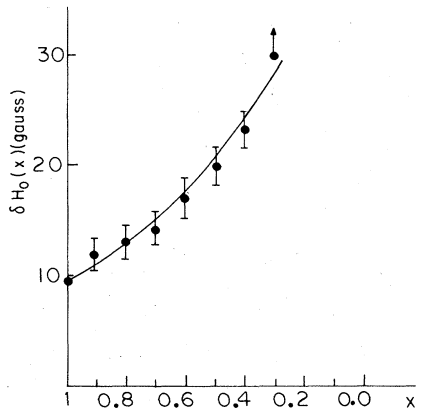

FIG. 2. Extrapolated zero-field linewidths of the I_1 ^{19}F resonance in $KMn_xMg_{1-x}F_3$, from the work of Borsa and Jaccarino (Ref. 6). The solid line was calculated from Eq. (6) of the text.

In view of the previous analysis, the behavior of $KNi_xMg_{1-x}F_3$ is quite intriguing. Because of the comparatively large exchange constant, the linewidth in pure[15,16] $KNiF_3$ [and also $\delta H(1)$] is actually smaller than in $KMnF_3$. The other factors that enter into Eq. (6) are mainly dependent upon the crystal structure which is identical in both systems. The failure to observe the I_1 and I_2 resonances in $KNi_xMg_{1-x}F_3$, is, therefore, somewhat puzzling. There exists some evidence[17] that a different percolation model may be necessary to interpret the experimental results in $KNi_xMg_{1-x}F_3$. One should notice that the analysis leading to Eq. (6) relies on the specification of what configuration of atoms actually constitutes a cluster of exchange coupled magnetic ions. In the model assumed to be valid for $KMn_xMg_{1-x}F_3$, two magnetic ions at nearest-neighboring sites are considered to belong to the same cluster independently of the occupancy of other neighboring sites. For a simple-cubic lattice this yields a percolation concentration[11] $x_p=0.311$ which appears to be in agreement with the experimental results in $KMn_xMg_{1-x}F_3$. For Ni^{2+} the situation may be entirely different. Since the ground state of this ion is ^3F, slight distortions from octahedral symmetry resulting from incomplete substitution of all six nearest-neighboring magnetic ions by nonmagnetic atoms can have a much larger effect than in Mn^{2+}, which has a half-filled shell with $L=0$. Although the electronic wave function at the ligands may not significantly vary by this process leaving the transferred hyperfine interaction unchanged, the wave function at the magnetic ion may be altered sufficiently in the case of Ni^{2+} to affect the exchange coupling with a neighboring ion. These arguments suggest that a more realistic model for $KNi_xMg_{1-x}F_3$ might result if one assumes that the exchange coupling between two nearest-neighboring Ni^{2+} ions depends upon the occupancy of other neighboring sites. Since quantitative calculations of superexchange are quite difficult, we decided to test a simple model based upon the following assumption: Two nearest-neighboring magnetic ions are considered as members of the same magnetic cluster only if their own nearest neighbors, along the line joining the two ions but in opposite directions, are also magnetic. We have performed preliminary Monte-Carlo simulations using this model, as well as, improved mean-field calculations. The percolation concentration, for example, appears to be considerably larger for this model than for conventional percolation. For a square lattice we obtained $x_p=0.73$ in-

stead of the value $x_p = 0.593$ valid for conventional site-dilution percolation in a square lattice.[11] It is worth pointing out that the variation of ordering temperature with concentration of magnetic ions, determined by NMR in $KNi_xMg_{1-x}F_3$,[17] actually displays a tendency towards a larger value of x_p with an apparent crossover at the lower temperatures. In addition to predicting a higher percolation concentration, the model outlined above would have the effect of increasing, in Eq. (6), the weight of sorted autocorrelation functions $\langle S_\alpha^{(r)}(\tau) S_\alpha^{(r)}(0) \rangle_{\text{av}}$ with smaller values of r. This would also lead to larger linewidths in $KNi_xMg_{1-x}F_3$ than in $KMn_xMg_{1-x}F_3$ for the same value of x and could probably also explain other differences between the behavior of both systems. Further work along these lines is currently in progress and will be reported in detail elsewhere.

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