Nuclear spin-lattice relaxation in ferrimagnetic clusters and chains: A contrast between zero and one dimensions

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Motivated by ferrimagnetic oligonuclear and chain compounds synthesized by Caneschi et al., both of which consist of alternating manganese(II) ions and nitronyl-nitroxide radicals, we calculate the nuclear spin-lattice relaxation rate $1/T_1$ employing a recently developed modified spin-wave theory. $1/T_1$ as a function of temperature drastically varies with the location of probe nuclei in both clusters and chains, though the relaxation time scale is much larger in zero dimension than in one dimension. $1/T_1$ as a function of an applied field in long chains forms a striking contrast to that in finite clusters, diverging with decreasing field like inverse square root at low temperatures and logarithmically at high temperatures.

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I. INTRODUCTION

Clusters of metal ions [1] serve to test the validity of quantum mechanical approaches at the nanometer scale. Among others a dodecanuclear manganese complex of formula $[\text{Mn}_{12} \text{O}_{12}(\text{CH}_3\text{COO})_{16}(\text{H}_2\text{O})_4]$ [2] and an octanuclear iron complex of formula $[\text{Fe}_8(\text{N}_3\text{C}_6\text{H}_{15})_6\text{O}_2(\text{OH})_{12}]^{8+}$ [3], which are hereafter abbreviated as $\text{Mn}_{12}$ and $\text{Fe}_8$, respectively, have been attracting considerable interest in this context. In these clusters, magnetic hysteresis curves are independent of temperature and consist of equally separated steps at sufficiently low temperatures [4–7], suggesting quantum tunneling of the magnetization. Both clusters have a magnetic ground state of total spin 10 and are often treated as a rigid spin-10 object with an Ising-type magneto-crystalline anisotropy. Such a phenomenological interpretation is indeed successful for the magnetic relaxation at sufficiently low temperatures and weak fields, but it masks individual internal structures of various magnetic clusters. Without a microscopic Hamiltonian, we could not essentially distinguish the $\text{Mn}_{12}$ and $\text{Fe}_8$ clusters. We cannot compare nanomagnets with bulk magnets in the same microscopic language until we describe them in terms of their constituent ion spins.

Thus motivated, several authors made an attempt to estimate exchange interactions in the $\text{Mn}_{12}$ and $\text{Fe}_8$ clusters calculating the low-lying energy spectra [8–10], magnetic susceptibilities [11], magnetization curves [12,13], inelastic neutron-scattering spectra [14], tunneling splittings [15], and nuclear spin-lattice relaxation rates [16]. The 8-spin modeling of the $\text{Mn}_{12}$ cluster [14,15] revealed the decisive role of multispin effects in magnetic tunneling, while the extended $ab\ initialo$ calculation [10] demonstrated the relevance of metal-ligand orbital hybridization to intramolecular exchange interactions. However, the predicted magnetic structures are rather controversial for both clusters. The $\text{Mn}_{12}$ cluster consists of eight Mn$^{3+}$ ions of spin 2 and four Mn$^{4+}$ ions of spin $\frac{3}{2}$, while the $\text{Fe}_8$ cluster contains eight Fe$^{3+}$ ions, both of which are coupled to each other through four kinds of exchange interactions (Fig. 1). Antisymmetric exchange interactions of the Dzyaloshinsky-Moriya type are also assumed to be relevant to the $\text{Mn}_{12}$ cluster [14]. There is no established way of assigning the magnetic anisotropy to each ion site. Thus, complicated intrachannel magnetic structures block our microscopic understanding of mesoscopic magnetism. Nanomagnets such as $\text{Mn}_{12}$ and $\text{Fe}_8$ have indeed been providing fascinating observations but are not necessarily suitable for a comparative study on zero- and one (or higher)-dimensional quantum magnetism.

![Figure 1](image_url)  
**FIG. 1.** Schematic plot of $\text{Mn}_{12}$ (a), $\text{Fe}_8$ (b), and $(\text{MnNIT})_6$ (c). Symmetry-in inequivalent sites are distinguishably drawn.
In order to reveal how paramagnetic spins grow into bulk magnets, we take a great interest in ferrimagnetic ring clusters \[17\] and chains \[18\] synthesized by Caneschi et al., both of which consist of manganese hexafluoroacetacetones (hfac) and nitronyl nitroxide radicals 2-R-4,4,5,5-tetramethyl-4,5-dihydro-1H-imidazolyl-1-oxyl (NIT-R) with R = phenyl (Ph), isopropyl (i-Pr), ethyl (Et), methyl (Me). The [Mn(hfac)NIT-\(\infty\)]\(_0\) cluster, hereafter abbreviated as (MnNIT)\(_0\), has antiferromagnetic exchange coupling between the Mn\(^{2+}\) ions of spin \(\frac{5}{2}\) and the radicals of spin \(\frac{1}{2}\) so as to exhibit a magnetic ground state of total spin 12 (Fig. 1). The [Mn(hfac)NIT-R]\(_\infty\) chain, hereafter abbreviated as (MnNIT)\(_\infty\), may be regarded as a one-dimensional analog of (MnNIT)\(_0\). Their simple magnetic structures, describable within isotropic exchange Hamiltonians \[17,18\], are suitable enough to compare oligonuclear ferrimagnets with those of one dimension in their intrinsic features.

The theoretical tool we employ here is a recently developed modified spin-wave theory, which is quite useful in understanding thermal \[19–21\] as well as ground-state \[22,23\] properties of various one-dimensional ferrimagnets. We inquire further into zero dimension and dynamic properties. Besides direct observations of resonant magnetization tunneling, nuclear-magnetic-resonance (NMR) measurements have extensively been performed for cluster magnets. \(^{1}\)H, \(^{2}\)D, \(^{13}\)C, and \(^{55}\)Mn NMR in Mn\(_{12}\) \[24–28\], \(^{1}\)H and \(^{2}\)D NMR in Fe\(_8\) \[29,30\], and \(^{1}\)H NMR in a hexanuclear copper complex \[31\] significantly contributed toward revealing the quantum dynamics of cluster magnets. As for the systems of Mn(hfac)NIT-R, we may consider NMR measurements using as probes \(^{1}\)H, \(^{13}\)C, \(^{19}\)F, and \(^{55}\)Mn nuclei. We calculate the nuclear spin-lattice relaxation rate \(1/T_1\) as a function of temperature, an applied field, and the location of probe nuclei.

**II. MODEL HAMILTONIAN**

\[
\mathcal{H} = J \sum_{i=1}^{N} (\mathbf{S}_i \cdot \mathbf{s}_i + s_i \cdot \mathbf{S}_{i+1}) - g \mu_B H \sum_{i=1}^{N} (s_i^z + S_i^z),
\]

where \(S = \frac{5}{2}\), \(s = \frac{1}{2}\), and we have set their \(g\) factors both equal to \(g\). Introducing bosonic operators for the spin deviation in each sublattice via \(S_i^+ = (2S - a_i^\dagger a_i)^{1/2} a_i\), \(S_i^- = S - a_i^\dagger a_i\), \(s_i^+ = b_i^\dagger (2s - b_i^\dagger b_i)^{1/2}\), \(s_i^- = -s + b_i b_i\), we expand the Hamiltonian with respect to 1/\(S\) as

\[
\mathcal{H} = -2SsJN + \mathcal{H}_1 + \mathcal{H}_0 + O(S^{-1}),
\]

where we assume that \(O(S) = O(s)\) and \(\mathcal{H}_i\) is the \(O(S^i)\) contribution. Considering the perturbational treatment of \(\mathcal{H}_0\) to \(\mathcal{H}_1\), we obtain the diagonal spin-wave Hamiltonian as

\[
\mathcal{H} = E_k + \sum_k \left[ \omega^- (k) a_k^\dagger a_k + \omega^+ (k) b_k^\dagger b_k \right] + O(S^{-1}),
\]

with \(E_k = -2SsJN + \sum_{i=1}^{N} E_i \omega_i^\pm (k)\) and \(\omega_i^\pm (k) = \sum_{i=1}^{N} \omega_i^\pm (k)\), where \(E_i\) and \(\omega_i^\pm (k)\) are the \(O(S^1)\) corrections to the ground-state energy and the dispersion relations, respectively. The dispersions of the linear spin waves, \(\omega_i^\pm (k)\), and the corrections due to the interactions between them, \(\omega_0^\pm (k)\), are, respectively, given by

\[
\omega_i^\pm (k) = (\omega_k \pm (S-s)J) \pm g \mu_B H,
\]

\[
\omega_0^\pm (k) = -2(S + J) \Gamma_1 \sin^2 (k/2) \omega_k - \frac{J \Gamma_2}{\sqrt{S}} \Omega_0 (\omega_k \pm (S-s)),
\]

where \(\omega_k = ([S-s]^2 + 4S s \sin^2 (k/2)]^{1/2}\), \(\Gamma_1 = -(1/2N) \sum_k [1 - (S + s)/\omega_k]\), and \(\Gamma_2 = (1/N) \sum_k (\sqrt{S} / \omega_k) \cos^2 (k/2)\). \(\Gamma_1\) is nothing but the quantum spin reduction \((1/N) \sum_k \langle a_i^\dagger a_i \rangle_{T=0} = (1/N) \sum_k \langle b_i^\dagger b_i \rangle_{T=0}\) and can analytically be evaluated as \(1/\sqrt{3} + 1/\sqrt{2} T - 7/24\) at \(N = 6\), which is slightly larger than the \(N \to \infty\) numerical estimate 0.106139 and thus suggests growing quantum fluctuations with decreasing system size. In Fig. 2 we plot the spin-wave dispersions together with quantum Monte Carlo calculations. The lower branch, which reduces the ground-state magnetization, is of ferromagnetic aspect exhibiting a quadratic dispersion at small momenta, whereas the upper branch, which enhances the ground-state magnetization, is of antiferromagnetic aspect being gapped from the ground state. The antiferromagnetic mode is remarkable for its \(O(S^0)\) quantum correction.

![FIG. 2. Dispersion relations of the linear (LSW) and interacting (ISW) spin waves, \(\omega_i^\pm (k)\) and \(\omega_0^\pm (k)\), at \(N = 32\) are also shown for reference.](image-url)
we obtain the optimum distribution functions as together with the trivial constraints \[ \sum \] with respect to \( P \) the Takahashi scheme [32] and enables us to investigate the dispersion relations are determined without modifying the original Hamiltonian (2.2) and then the Lagrange multiplier is introduced so as to construct a reliable thermodynamics, which is essentially different from the Takahashi scheme [32] and enables us to investigate much wider temperature range. Otherwise the Schottky peak of the specific heat, for instance, cannot be reproduced at all [19]. In this context, a mixed Bose-Fermi representation of spin operators [33] may be another useful scheme to thermal calculations.

The magnetic susceptibility is calculated as

\[ \chi = \frac{(g\mu_B)^2}{3k_B T} \sum_{\sigma=\pm} \bar{n}_k^\sigma (\bar{n}_k^\mp + 1), \] (3.4)

and is shown in Fig. 3, together with quantum Monte Carlo calculations at \( N = 32 \), which are almost the long-chain-limit behavior. Modified spin-wave calculations well agree to the numerical findings, covering the low-temperature region to be hardly reached numerically. The decreasing behavior turns increasing for \( k_B T \gtrsim 3J \). A minimum in the susceptibility-temperature product is one of the most remarkable features of ferrimagnets. The ferromagnetic and antiferromagnetic terms \( \bar{n}_k^\pm (\bar{n}_k^\mp + 1) \) in Eq. (3.4), respectively, contribute increasing and decreasing behaviors with increasing temperature. Since the ferromagnetic (antiferromagnetic) features are predominant for \( S > 2s \) \( (S < 2s) \) [34], the present cases with \((S, s) = (\frac{3}{2}, \frac{1}{2})\) are rather biased ferromagnetically. In order to evaluate the exchange coupling constants in the Mn(hfac)NIT-R systems, we further compare quantum Monte Carlo calculations with experimental findings. The susceptibility measurements on Mn(hfac)NIT-Ph at \( H = 0.5 T \) [17] and those on Mn(hfac)NIT-Pr at under no field [18] are also plotted in Fig. 3, taking \( J/k_B \) to be 370 K and 360 K, respectively. Carrying out semi-classical calculations, Caneschi et al. [18] estimated \( J/k_B \) for Mn(hfac)NIT-Pr as 474.5 K \(( s = 3, \frac{1}{2}) \), 373.3 K \(( s = 4, \frac{1}{2}) \) and 311.8 K \(( s = 5, \frac{1}{2}) \). However, smaller values are obtained through a Fischer’s model [35]. As for Mn(hfac)NITPh, no quantitative assignment of \( J \) has been given so far. The present estimates will contribute toward establishing the standard.

**IV. NUCLEAR SPIN-LATTICE RELAXATION**

NMR measurements on isotropic magnets are necessarily performed with an applied field. An induced Zeeman energy gap is usually smaller than the exchange interaction but larger than the nuclear energy scale: \( 10^2 h\omega_N \ll 10^2 g\mu_B H \ll J \). Considering the electronic-nuclear energy-conservation requirement, the Raman process should play a leading role in the nuclear spin-lattice relaxation [36]. The Raman relaxation rate is generally given by

\[ \frac{1}{T_1} = \frac{4\pi h(g\mu_B gN)^2}{\sum_n e^{-E_n/k_BT}} \sum_{n,m} e^{-E_m/k_BT} \times |\langle m | \sum_i (A_i^z S_i^z + A_i^z S_i^z) | n \rangle|^2 \delta(E_m - E_n - \hbar\omega_N), \] (4.1)
where $A_{ij}^z$ and $a_{ij}^z$ are the dipolar coupling constants between the nuclear and electronic spins in the $j$th unit cell, $\omega_N \equiv \gamma_N H$ is the Larmor frequency of the nuclei with $\gamma_N$ being the gyromagnetic ratio, and the summation $\sum_n$ is taken over all the electronic eigenstates $|n\rangle$ with energy $E_n$. Taking account of the significant difference between the electronic and nuclear energy scales, the relaxation rate (4.1) is expressed in terms of modified spin waves as

$$\frac{1}{T_1} \simeq \frac{2\hbar (g \mu_B \gamma_N)^2}{N} \sum_{\sigma=\pm} \left[ \sum_k |A_{ij}^z \psi^{\sigma}(k) - a_{ij}^z \psi^{\sigma}(k)|^2 \bar{n}_k^\sigma (\bar{n}_k^\sigma + 1) \rho^\sigma(k) \right.$$

$$+ \left. \sum_k |A_{2ij}^z \psi^{\sigma}(k) - a_{2ij}^z \psi^{\sigma}(k)|^2 \bar{n}_k^\sigma (\bar{n}_k^\sigma + 1) \rho^\sigma(k) \right], \quad (4.2)$$

where $\sum_k$ denotes the limited summation $\sum_k - \sum_{k=0,\pi}$, $\psi^\sigma(k) = (S + s)/2[(S - s)^2 + 4s \sin^2(k/2)]^{1/2} + \sigma/2$, and $A_{ij}^z$ and $a_{ij}^z$ are the Fourier transforms of the coupling constants, whose $k$ dependences are hereafter assumed to be negligible. A contrast between zero and one dimensions lies in the spectral density $\rho^\sigma(k)$, which originates in the energy-conservation requirement $\delta(E_m - E_n - \hbar \omega)$ in Eq. (4.1). In the thermodynamic limit $N \to \infty$, $\rho^\sigma(k)$ is definitely the differential coefficients of the dispersion relations, while for small clusters, it is approximately replaced by the difference quotients:

$$\rho^{\pm}(k) = \frac{1}{[d\omega^{\pm}(k)/dk]_{k=k_0}} \quad \text{for } N \to \infty, \quad (4.3a)$$

$$\rho^{\pm}(k) \simeq \frac{2\pi/N}{[\omega^{\pm}(k) - \omega^{\pm}(k - 2\pi/N)]} \quad \text{for } N = O(1), \quad (4.3b)$$

where $k_0$ as a function of $k$ is given by $\omega^{\pm}(k_0) - \omega^{\pm}(k) - \hbar \omega = 0$. If we further process Eq. (4.3a) assuming the predominance of $k \simeq 0$ contributions [37] in integrating Eq. (4.2), which is well justified unless temperature and an applied field are sufficiently high and strong, respectively, we obtain an expression

$$\rho^{\pm}(k) \simeq \frac{1}{2\nu \sqrt{k^2 + \hbar \omega / v}} \quad \text{for } N \to \infty, \quad (4.4)$$

where $v = [Ss - (S + s)T_1 + \sqrt{SsT_2}/J] / (S - s)$ is the curvature of the dispersion relations at small momenta. Equation (4.4) is in contrast with Eq. (4.3b) in that it depends on an applied field.

Another consideration should be directed to the modified spin-wave scheme in calculating the relaxation rate. For isotropic ferrimagnets with gapless excitations, the constraint (3.1) works so well as not only to suppress the thermal divergence of the boson number but also to give a precise description of the low-temperature thermodynamics [38]. On the other hand, once a field is applied and a gap $\Delta$ opens in the electronic energy spectrum, the boson number should exponentially decreases as $\propto e^{-\Delta/k_B T}$ at low temperatures, whereas the constraint still keeps it finite even at $k_B T < g \mu_B H$. Then we adjust Eq. (3.1) to the present situation as

$$(S + s) \sum_k \sum_{\sigma=\pm} \bar{n}_k^\sigma = (S + s)Ne^{-\Delta/k_B T}. \quad (4.5)$$

This condition smoothly turns into Eq. (3.1) as $H \to 0$ and the modification is essentially restricted to the sufficiently low-temperature region $k_B T \lesssim \Delta \lesssim 10^{-2} J$.

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**FIG. 4.** The interacting-spin-wave calculations of the temperature dependence of the nuclear spin-lattice relaxation rate with varying location of the probe nuclei at $N = 6$ (the upper three) and $N = \infty$ (the lower three).
spin relaxation, in other words, of \( \sigma \) nuclear and electronic spins. Figure 4 shows \( 1/T \) which can be described by a parameter \( r \). Considering the predominance of the electronic excitations \( \psi \), the \( \sigma \) excitation mode hardly mediates the nuclear spin relaxation, in other words, the electronic excitations of \( \sigma \) mode are invisible to the nuclear spins. The temperature dependence of \( 1/T_1 \) is indeed of antiferromagnetic (ferromagnetic) aspect at \( r = 1/5 \) (\( r = 5 \)), while otherwise it is of mixed aspect. If the probe nuclei are located as \( r \approx 1/5 \), the relaxation rate is extremely small. These calculations can be observed by taking different kinds of nuclei as probes, such as \(^1\)H, \(^{13}\)C, \(^{19}\)F, and \(^{55}\)Mn in the present systems. Equation (4.2) is still valid for \( r \rightarrow \infty \), which corresponds to \(^{55}\)Mn NMR.

Figure 5 shows \( 1/T_1 \) as a function of an applied field. There appears a clear contrast between zero and one dimensions, where \( 1/T_1 \) is saturated and diverging, respectively, with decreasing temperature and field, as long as the ferromagnetic spin waves are visible to the nuclear spins. In finite clusters, any field dependence of \( 1/T_1 \) is necessarily attributed to \( \tilde{n}_k^\pm \), which are simply exponential with respect to \( H \), whereas in long chains, \( 1/T_1 \) is more varied with a field, depending on it through both \( \tilde{n}_k^\pm \) and \( \rho^\pm(k) \). Therefore, unless the Zeeman energy becomes comparable to the exchange interaction, \( 1/T_1 \) exhibits little field dependence in finite clusters. In order to bring out their characteristic field dependences more quantitatively, we plot \( 1/T_1 \) as a function of \( \sqrt{H} \) in Fig. 6. \( \tilde{n}_k^\pm \) and \( \rho^\pm(k) \) are both peaked at \( k = 0 \). More and more weight centers on \( k = 0 \) with decreasing temperature and field for \( N \rightarrow \infty \) in particular. Therefore, at low temperatures, the \( N \rightarrow \infty \) \( k \) integration in Eq. (4.2) may approximately be replaced by the \( k = 0 \) contribution, which is in proportion to \( 1/\sqrt{H} \). Thus, as long as an applied field is moderate, a \( 1/\sqrt{H} \)-linear behavior is observed at low temperatures. With increasing temperature it turns logarithmic, sloping more gently, due to the \( k \)-integration effect. Under strong fields, they are all masked behind the overwhelming Zeeman term \( \propto e^{-g\mu_B H/k_B T} \) coming from \( \tilde{n}_k^- \).

At \( r = 1/5 \), where only the antiferromagnetic spin
waves are active for $1/T_1$, the nuclear spins exhibit extremely slow dynamics. Since $n^+_n$ is not peaked even at low temperatures, there appears no $1/\sqrt{H}$ dependence. With increasing temperature and field, the antiferromagnetic excitation branch lowers in energy and thermally assists the relaxation, ending up with increasing $1/T_1$. A minimum of $1/T_1$ as a function of $H$ can be observed only when the ferromagnetic spin waves are almost off. In finite clusters, the discrete spectrum may in principle lead to $1/T_1$ oscillating as a function of $H$.

### V. CONCLUDING REMARKS

Motivated by inorganic-organic hybrid compounds, $[\text{Mn(hfac)NIT-Ph}]_6$ and $[\text{Mn(hfac)NIT-R}]_\infty$, we have demonstrated model calculations of the low-energy spin dynamics in ferrimagnetic clusters and chains. Temperature dependence of $1/T_1$ drastically varies with the location of the nuclei in both compounds, though the relaxation time scale is much larger in zero dimension than in one dimension. There are special points for the nuclei, characterized as $(d_s/d_s)^3 \sim s/S$, where the nuclear spin relaxation can hardly be assisted by the low-lying ferromagnetic excitations of the electronic spins and therefore extremely slow dynamics is observed.

Field dependence of $1/T_1$ in long chains forms a striking contrast to that in finite clusters, diverging with decreasing field at low temperatures. The present observations should be distinguished from the $1/\sqrt{H}$ or $\ln(1/H)$ dependence of diffusion-dominated dynamics [39,40], which originates from transverse spin fluctuations and distinctly appears at high temperatures. In the vicinity of the special points of $(d_s/d_s)^3 \sim s/S$, a minimum of $1/T_1$ as a function of $H$ can be observed.

Besides the Mn(hfac)NIT-R systems, there are a series of ferrimagnetic bimetallic chain compounds $M\text{Cu(pba)}(\text{H}_2\text{O})_3\cdot n\text{H}_2\text{O}$ ($M = \text{Mn, Ni}$; pba = 1,3-propylenebis(oxamato)) [41] and $M\text{Cu(pbaOH)}(\text{H}_2\text{O})_3\cdot n\text{H}_2\text{O}$ ($M = \text{Fe, Co, Ni}$; pbaOH = 2-hydroxy-1,3-propylenebis(oxamato)) [42], which are also describable by the Hamiltonian (2.1). Since their exchange coupling constants are much smaller than those of the Mn(hfac)NIT-R systems, they are complementary in field-applied measurements. We hope that the present calculations will stimulate further experimental explorations into quantum dynamics on the way from zero- to one-dimensional magnets. Nuclear spin-lattice relaxation-time measurements on $[\text{Mn(hfac)NIT-Ph}]_6$ and $[\text{Mn(hfac)NIT-R}]_\infty$ are strongly encouraged.

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