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Dynamic spin fluctuations in the molecular ferromagnet (DMeFc)(TCNE)

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The static and dynamic magnetic properties of the molecular ferromagnet decamethylferrocenium tetracyanoethenide (DMeFc)(TCNE) are studied via the muon-spin-relaxation technique. Spontaneous order is observed in the ferromagnetic ground state below 5 K, while the muon-spin-relaxation rate in the paramagnetic phase displays a gradual variation with temperature, indicating that a slowing down of spin fluctuations occurs over a wide temperature range. The temperature dependence of spin fluctuations between 8 and 80 K shows the thermally activated behavior expected in a spin chain with Ising character.

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I. INTRODUCTION

There have been extensive studies of the mixed-stack electron transfer salt decamethylferrocenium tetracyanoethenide[1–6] (DMeFc)(TCNE) since the first report of molecular ferromagnetism in this system.[2] This system has a stack of DMeFc (donor) and TCNE (acceptor) molecules forming a one dimensional structure, as shown in Fig. 1. The Fe^{3+} ion in DMeFc is in a low-spin state with $S=1/2$, which gives an anisotropic and large g value[2] to the unpaired spin of the DMeFc molecule, while an electron transferred to TCNE ($S=\frac{1}{2}$) with an isotropic g value also contributes to the magnetism. Bulk measurements, such as magnetization,[2,3] specific heat,[4] and neutron diffraction,[5] showed that the system exhibits primarily one-dimensional (1D) behavior at high temperatures, and undergoes a phase transition at $T_c=4.8$ K to a 3D ferromagnetic ground state. A substantial anisotropy of interchain and intrachain magnetic exchange interactions in this system was confirmed by susceptibility studies.[6] So far, measurements focused on static bulk properties, while much less work was done concerning dynamic spin fluctuations in this quasi-1D ferromagnet.

Dynamic magnetic properties of 1D magnets are particularly interesting, owing to the existence of strong short-range spin correlations and a broad critical region.[7] Extensive theoretical studies were performed[7,8] to explain various elementary magnetic excitations, both linear and nonlinear. On the other hand, direct observations of spin fluctuations, by neutron scattering or NMR, have been performed so far only in a limited number of systems,[8,9] most of which have antiferromagnetic and/or Heisenberg interactions. To verify these model excitations in various real systems, we study dynamic properties in the present molecular ferromagnet, which has a strong 1D character with a substantial Ising anisotropy.

We report muon-spin-relaxation (μSR) studies[10,11] in both ferromagnetic and paramagnetic states of (DMeFc)(TCNE). The present experiments clearly show the existence of slow dynamics over a wide temperature range in this molecular ferromagnet, due to the suppression of long-range order in the quasi-1D system. To our knowledge, this is the first reported study of slow magnetic fluctuations in 1D spin systems performed using the μSR technique. This work adds an interesting aspect to existing μSR studies of magnetism in organic systems.[12,13]

II. EXPERIMENTAL METHOD AND SPECIMENS

In long-range-ordered magnetic systems, a spontaneous magnetization M_s appears below T_c. Zero-field (ZF) μSR provides information about M_s via a Larmor frequency ω_μ of muon spins around an internal magnetic field $B_{\rm int}$, $M_s \propto \omega_\mu = \gamma_\mu B_{\rm int}$, where the muon gyromagnetic ratio $\gamma_\mu = 85.16$ kHz/G. In the paramagnetic phase above T_c, M_s vanishes and is replaced by dynamic and spatial spin correlations which decay at long time and long distance: the average correlation time τ and correlation length ξ decrease with increasing temperature.

Dynamic effects are detected via muon-spin depolarization in time. For fast fluctuations (in the narrowing limit $\gamma_\mu \Delta \tau \ll 1$, where Δ denotes a typical value of $B_{\rm int}$), the muon-spin polarization function can be expressed approximately as

$$P_z(t) = \exp[-(t/T_1)^p], \tag{1}$$

where T_1 is the relaxation time and the power p is of order unity. If the spin correlation is characterized by a single correlation time τ, $\mathbf{S}(t)\cdot\mathbf{S}(0) \propto \exp(-t/\tau)$, T_1 is determined by[14]

$$T_1^{-1} = C\gamma_\mu^2 \Delta^2 \tau. \tag{2}$$

The coefficient C depends upon the instantaneous field distribution $\rho(B_{\rm int}/\Delta)$. For instance, $C=2$ for a Gaussian-distributed ρ and $C=4$ for a Lorentzian ρ. With the application of a longitudinal field (LF, where $\mathbf{H}_L$ is parallel to the initial muon spin), T_1 can be expressed as

$$T_1^{-1}(H_L) = C\gamma_\mu^2 \Delta^2 \tau/(1+\omega_L^2\tau^2), \tag{3}$$

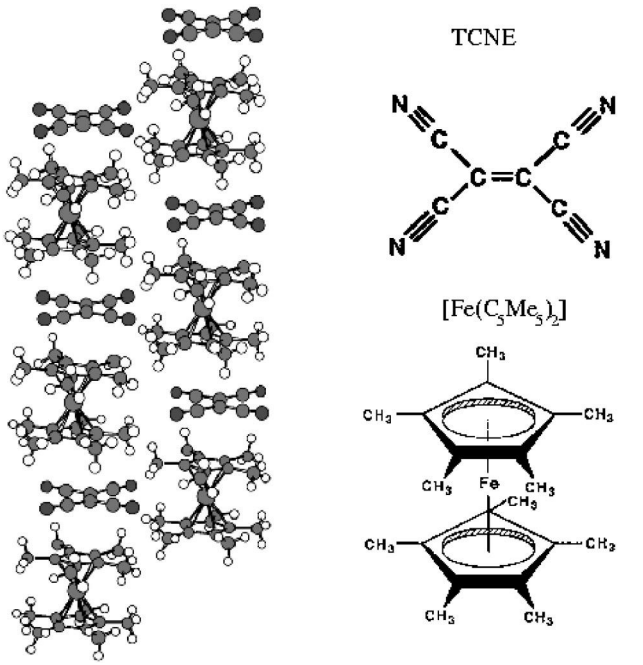

FIG. 1. Crystal structure of decamethylferrocenium tetracyanoethenide (DMeFc) (TCNE). The left figure shows a one dimensional stack of DMeFc donors and TCNE acceptors, while the right figures show molecular structures of these TCNE and DMeFc. The Fe^{3+} in DMeFc is in an $S=\frac{1}{2}$ low-spin state.

where $\omega_L=\gamma_\mu H_L$. The power p also depends on $\rho(B_{\rm int}/\Delta)$: $p=1$ for the Gaussian distribution and $p=1/2$ for the Lorentzian distribution, respectively.

Our sample of (DMeFc)(TCNE) was prepared as previously described.[3] Two pellets, each 20 mm in diameter and 3 mm thick, were pressed at 10 kbar. The specimens were sealed in argon gas before the experiments and in helium gas during the μSR measurements to avoid air contamination. The μSR experiments were performed using the positive muon beam at TRIUMF, with the initial muon spin along the beam direction and normal to the sample plane surface. The temperature dependence of the magnetic properties was studied in sample No. 1, while the field dependence of the dynamic behavior in the paramagnetic state was examined in sample No. 2. We found that the μSR spectra in both samples are essentially the same at 7 K, whereas the relaxation rate around 10 K of one sample was close to that at 11 K of the other, indicating a small difference in sample quality and/or some systematic uncertainty in measurements.

III. EXPERIMENTAL RESULTS

Figure 2(a) shows the μSR time spectra observed at 3 K. A dip visible at 0.1 μs in the ZF-μSR spectrum is characteristic of static magnetic fields. The absence of long-lived oscillations, on the other hand, indicates a broad distribution of $B_{\rm int}$. This feature is in contrast with our previous observation in the organic ferromagnet β phase *p*-nitrophenylnitronylnitroxide,[15] where a well-defined oscillation was observed below T_c, which allowed a precise determination of $M_s(T)$.

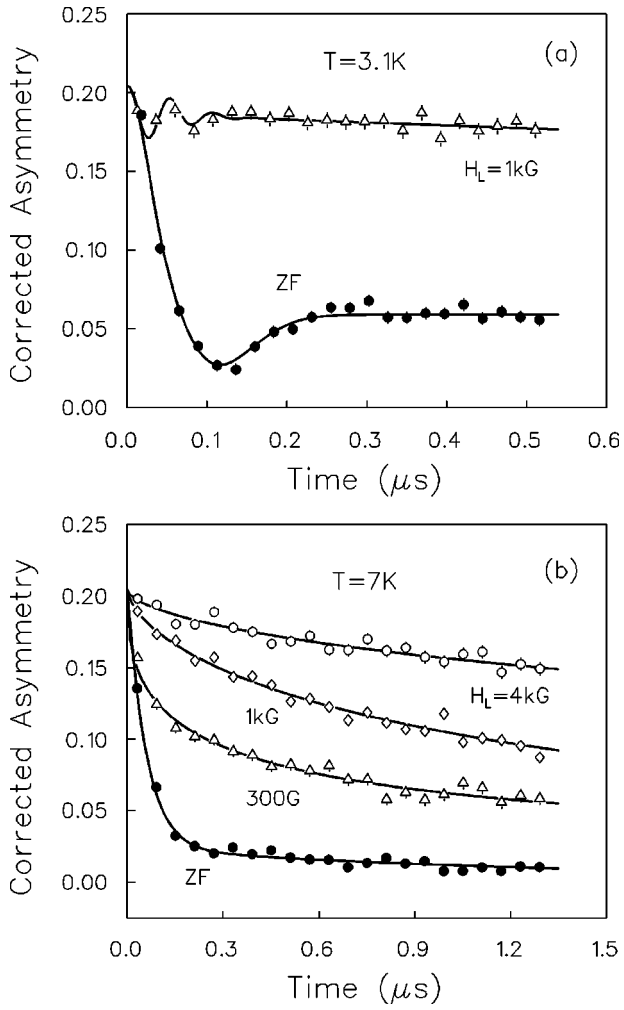

FIG. 2. Zero-field and longitudinal-field spectra observed in (DMeFc)(TCNE) at 3 K (a) and 7 K (b). The muon decay asymmetry is shown after a correction related to solid angle factors and backgrounds. A spontaneous order, with a broad internal field distribution, appears at 3 K, while a dynamic behavior is seen at 7 K.

The ZF-μSR spectrum at 3 K can be described by a generalized Kubo-Toyabe (KT) function, which is appropriate for static random fields,

$$P_z^{\rm KT}=\frac{1}{3}+\frac{2}{3}[1-(\gamma_\mu\Delta t)^\beta]\exp[-(\gamma_\mu\Delta t)^\beta/\beta], \qquad (4)$$

where β is an adjustable parameter: $\beta=2$ for the Gaussian ρ and $\beta=1$ for the Lorentzian ρ, respectively. The best fit gives $\Delta=170$ G and $\beta=1.5$. Since Δ is significantly larger than the typical nuclear dipolar fields of several G, the observed internal magnetic field must be due to the electron magnetism. A reasonably large Δ is also consistent with a dense spin structure in (DMeFc)(TCNE). The value of β indicates that $\rho(B_{\rm int}/\Delta)$ lies somewhere between Gaussian and Lorentzian distributions. When applying a longitudinal field of 1 kG ($H_L/\Delta\approx 6$), the resultant local field (the vector sum of $\mathbf{B}_{\rm int}$ and $\mathbf{H}_L$) becomes essentially parallel to the initial muon spin, leading to a dramatic recovery of the muon-spin polarization, as shown in Fig. 2(a). The observed long-time

asymmetry of about 86% also lies in between the 94% and 79% expected for the Gaussian and Lorentzian field distributions, respectively.

Ferromagnetic order in the (DMeFc)(TCNE) system was well established by bulk measurements;[1–6] a spontaneous magnetization and hysteretic behavior are both seen below 4.8 K. The broad field distribution observed in the present study is thus *not* attributed to the intrinsic randomness of spin configuration, such as in spin-glass systems. The plausible origins for observed random fields are either multiple muon locations within the (DMeFc)(TCNE) lattice or the multidomain structure of a polycrystalline specimen, or both.

Generally, $B_{\rm int}$ is the sum of fields from dipoles both inside and outside a Lorentz sphere. The former contribution is sensitive to the muon location: one expects a large inhomogeneous field when the fields vary significantly from site to site (multiple muon sites). The latter effect is nontrivial when a net magnetization is present, as in each domain of a ferromagnet, where the field inhomogeneity depends on both the local-domain boundary and the nearby domain structure (the whole system appears somewhat like a macroscopic spin glass). For instance, the demagnetization of $4\pi NM$ varies from domain to domain with the variation of the demagnetization factor N from 0 to 1 (multiple domains). The magnitude of these macroscopically random fields is thus expected to be several times the magnetization M. From the saturation moment of 1.63×10^4 emu G/mole,[2] we estimate $M=40$ G, which is indeed comparable with $\Delta=170$ G. We see that even though the ferromagnetic spins are homogeneous in each domain, the magnetic fields "seen" by each muon could have a broad distribution.

Above 5 K, the absence of a dip in the ZF spectra indicates the disappearance of the static field, and hence a ferromagnetic-paramagnetic transition. Figure 2(b) shows the ZF and LF spectra obtained at 7 K. All spectra display a monotonic muon-spin depolarization, revealing the fluctuating nature of the system. When the temperature exceeds 20 K, fast fluctuations lead to a small T_1^{-1}, and the (background) nuclear dipolar relaxation becomes significant. To remove such effects, we apply $H_L=100$ G, significantly larger than the static nuclear dipolar fields of about 4 G. Since $H_L=100$ G has a negligible effect on fast fluctuations [$H_L\tau\ll1$, cf. Eq. (3)], the LF relaxation rate reflects the fluctuation rate at high temperature.

Both the ZF and LF spectra above T_c are fit to Eq. (1). We found that $p\approx0.7$ and was only weakly temperature dependent. This result seems reasonable, since the static field profile falls between the Gaussian and Lorentzian distributions (where $p=1$ and $\frac{1}{2}$, respectively). The value of T_1 reflects the typical fluctuation rate and can be determined accurately when the muon-spin depolarization occurs in a μSR time window of 10 μs.

We examined the detailed field dependence of the relaxation rate in sample No. 2 at 7 and 10 K, as shown in Fig. 3. Since the fluctuation rate at these temperatures is small, significant changes in the μSR spectra were observed with H_L being on order of 1 kG. The curves are fits according to Eq. (3). The data agree fairly well with Eq. (3) over the decade of change in T_1^{-1}. We recall that a sufficiently large external field may actually induce a static field above T_c, and therefore enhance the relaxation rate (cf. Fig. 5 in Ref. 14). Magnetization measurements in (DMeFc)(TCNE) show that the effective T_c rises to 7 K under an external field of 2 kG.[3] Hence Eq. (3) may be inappropriate under a large LF.

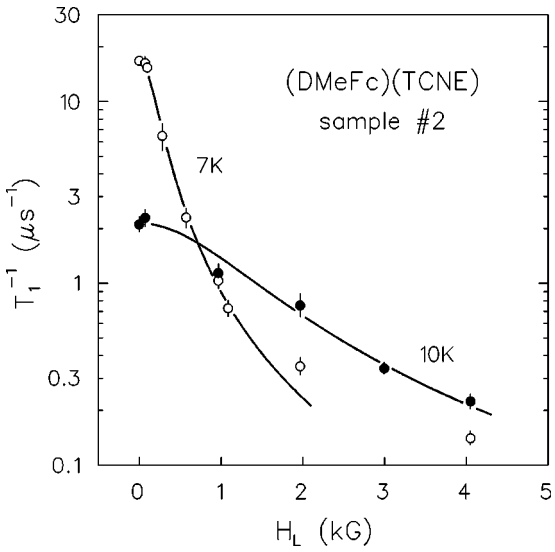

FIG. 3. Longitudinal-field dependence of the μSR relaxation rate at 7 and 10 K in sample No. 2. Curves are fits to Eq. (3).

The spin correlation time τ can be determined from the field dependence of T_1^{-1}, as demonstrated in Fig. 3. We find $\tau=5\times10^{-8}$ sec at 7 K and $\tau=9\times10^{-9}$ sec at 10 K, respectively. Substituting these values of τ and $T_1^{-1}(H_L=0)$ into Eq. (2), and using $C=3$, we obtained $\Delta=130$ G at 7 K and $\Delta=110$ G at 10 K, which agree reasonably well with the static internal field of 170 G derived at 3 K. The consistency of Δ obtained at different temperatures confirms the applicability of Eqs. (2) and (3) to the present system above 7 K.

Figure 4 shows the temperature dependence of relaxation rate observed in the paramagnetic phase of sample No. 1. The open circles represent the results obtained from ZF measurements for large T_1^{-1}, and solid circles those obtained by LF measurements ($H_L=100$ G) for small T_1^{-1}. Using $\Delta=120$ G and $C=3$ in Eq. (2), we also show the correlation time in Fig. 4. We find that τ decreases by three orders of magnitude when the temperature rises from 6 to 80 K.

In a 3D ordered system, the critical increase in τ occurs in only a small temperature region, within a percent of the critical temperature T_c [cf. NMR measurements in antiferromagnetic MnF_2 (Ref. 16)]. At higher temperatures, τ approaches a finite value, which is determined by exchange narrowing.[17] A broader critical reagion above T_c has been observed in disordered spin glass AuFe, CuMn and AgMn systems,[14,18] where τ decreases by more than two orders of magnitude with increasing temperature from T_c to $2T_c$, and saturates above $2T_c$.

In (DMeFc)(TCNE), the behavior of $\tau(T)$ is significantly different. For very slow fluctuations, i.e., $\tau>10^{-7}$ sec or $\gamma_\mu\Delta\tau>1$, the narrowing-limit formula Eq. (2) becomes no longer valid. It is therefore difficult to determine the behav-

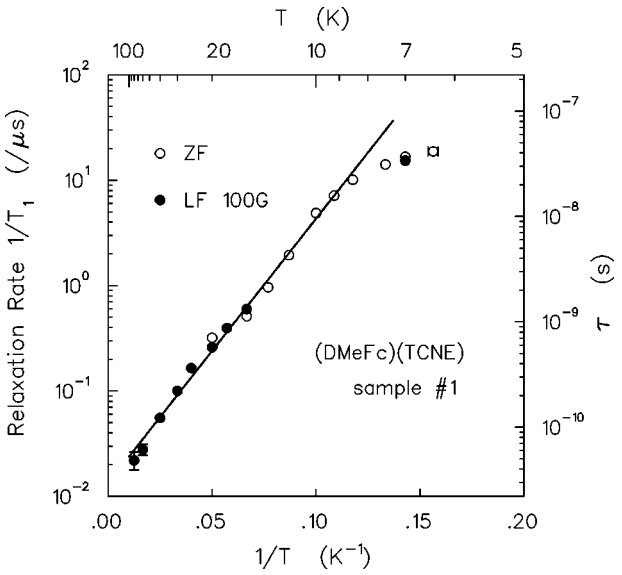

FIG. 4. Relaxation rate and correlation time vs reciprocal temperature in the paramagnetic phase of sample No. 1. The solid line shows a fit to Eq. (5), with an activation energy of $E_a/k_B = 57.8$ K.

ior of τ when the temperature is very close to T_c. Significant changes in τ are observed above 10 K; we find that τ decreases more than two decades between 10 and 80 K (i.e., $2T_c$ and $16T_c$). The data between 8 and 80 K can be described by thermally activated behavior, as demonstrated by the solid line in Fig. 4:

$$\tau(T) = \tau_\infty \exp(E_a/k_B T). \tag{5}$$

We extract activation energy of $E_a/k_B = 57.8$ K and $\tau_\infty = 5 \times 10^{-11}$ sec. Figure 4 clearly demonstrates that the critical slowing down of spin dynamics in the (DMeFc)(TCNE) system gradually develops over a wide temperature range.

IV. DISCUSSIONS AND CONCLUSIONS

When there exist mechanisms which suppress long-range order, the 3D ordering temperature T_c may be much lower than J/k_B, leading to the formation of short-range spin correlations between T_c and J/k_B. Consequently, slow fluctuations are expected to occur in a broad temperature range above T_c for correlated spins. In a geometrically frustrated spin system $SrCr_8Ga_4O_{19}$,[19] for example, this effect results in a variation of the muon-spin-relaxation rate over a wide temperature range above the spin freezing temperarture T_g inferred from the cusp in the ac susceptibility. The slow spin dynamics observed in (DMeFc)(TCNE) are most likely related to the quasi-1D nature of the system. For instance, susceptibility measurements[2,3] show a small ratio of T_c to the Curie-Weiss temperature, $T_c/\Theta = 0.16$, and specific-heat measurements[4] show that only 4% of the total entropy is involved in the 3D ordering.

The spin Hamiltonian of the system has been treated as a spin-$\frac{1}{2}$ Heisenberg chain with only nearest-neighbor interaction and a uniaxial anisotropy[4,6]

$$\mathcal{H} = -2J \sum_i \{S_z^i S_z^{i+1} + \epsilon(S_x^i S_x^{i+1} + S_y^i S_y^{i+1})\}, \tag{6}$$

where $J > 0$. A significant anisotropy of $\epsilon = 0.35$, due to the anisotropic Lande g value of the DMeFc ion,[2,4,6] indicates an Ising-like character of the present system.

To understand our experimental results in the framework of the model Hamiltonian [Eq. (6)], we can compare the spin dynamics of a Heisenberg chain (HC) with those of an Ising chain (IC). The dynamic spin correlation time is closely related to the spatial spin correlation length. We may write $\tau = \xi/v$, with v referring to the typical propagation velocity of quasiparticles. In a HC, the low-lying excitations are quasi-spin-waves with wavelengths shorter than ξ. At low temperatures,[20] $\xi \sim J/k_B T$ and $v \sim J/\hbar$. Then $\tau \sim \hbar/k_B T$. When a small anisotropy ($\delta = |1-\epsilon| \ll 1$, which may be due to either an intrinsic anisotropy or applied external field) is introduced to the HC, there exists another type of elementary excitation—solitons. Since δ is small, the solitons may extend over several lattice units (broad solitons). This model yields[21] that $\tau \sim \exp(E_s/k_B T)$, with soliton energy $E_s \sim J\sqrt{\delta}$.

In an IC, the basic excitations are the domain walls (narrow solitons, due to strong anisotropy). The dynamics in an antiferromagnetic (AFM) IC was first discussed by Villain,[22] who showed that $\xi \sim \exp(J/k_B T)$ and $v \sim \epsilon J/\hbar$. In the case of a ferromagnetic (FM) IC, the spin dynamics is quite different since the domain walls are static.[23] In order to move a wall, a biaxial anisotropy is required.[24] Thus the dynamics of a FM IC are essentially characterized by thermally activated processes. Since the energy required to flip spins within a pair of walls is $2J$, τ can be expressed as Eq. (5) with $E_a = 2J$.

We see that the slowing down of spin fluctuations with decreasing temperature is significantly distinguishable in HC versus IC systems, and in FM versus AFM systems. With the strongest correlation among the spins, the FM IC displays the most significant slowing down of spin fluctuations among the various 1D spin chains. The thermally activated behavior of τ observed in (DMeFc)(TCNE), with a large E_a, indicates that the present molecular ferromagnet has an Ising character. Using $E_a = 2J$, we obtain $J/k_B = 28$ K, consistent with $J/k_B \approx 35$ K estimated from the specific-heat measurements for $T > T_c$.[4]

We now compare our observed value of τ_∞ with various magnetic chains. At high temperature, $\xi \sim 1$ and $\tau \sim 1/v$. Then, in a HC, τ_∞ corresponds to the exchange narrowing of $\hbar/J$, while in an AFM IC, a larger $\tau_\infty \sim \hbar/(\epsilon J)$ is expected since the dynamics of S_z are determined by the transverse exchange interaction ϵJ. (It is noteworthy that a strict Ising Hamiltonian with $\epsilon = 0$ does not contain any dynamics.) In a FM IC, since the propagation of walls is forbidden, the slower diffusion process may lead to even larger τ_∞. Indeed, the experimental value of τ_∞ is found to be more than two orders of magnitude larger than $\hbar/J = 3 \times 10^{-13}$ sec.

We have observed slow spin dynamics over a wide temperature range in the paramagnetic phase of the molecular ferromagnet (DMeFc)(TCNE). The current results are very similar to previous Mössbauer measurements[25] in the FM IC $FeCl_2(NC_5H_5)_2$, where the authors found that $E_a \approx 2J$ and

τ_∞ is about two orders of magnitude greater than $\hbar/J$. The large values of E_a and τ_∞ deduced from the present μSR studies indicate that (DMeFc)(TCNE) has a quasi-1D spin structure with Ising character. This feature is consistent with previous bulk measurements.[4,6] For instance, specific-heat studies[4] showed a broad maximum of C_M at 15 K, expected for an Ising rather than a Heisenberg-like 1D ferromagnet. Below T_c, C_M varies as $\exp(-\Delta/k_BT)$ with $\Delta/k_B=22$ K. This gap may be related to the strongly anisotropic nature of the present system.

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