Diffusive Solitons in Impure Sine-Gordon Chains: Experimental Evidence in $(CD_3)_4NMn_{(1-c)}Cu_cCl_3$

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From nuclear relaxation-time measurements, experimental evidence is given that the magnetic solitons behave diffusively in the chains of $(CD_3)_4NMnCl_3$ doped with copper. Some neutron inelastic-scattering results are also presented, which support this analysis.

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It is clear nowadays that the concept of soliton is of great importance in many areas of physics. An essential problem concerns the dynamical behavior of these quasiparticles, in particular when they are subjected to perturbations and/or to interactions with a heat bath. Theoretical descriptions[1,2] and computer simulation studies[3] have recently been published on this problem. We present here the first results of an experimental study of a well-defined soliton regime perturbed by impurities.

As we have recently shown, the quasi one-dimensional (1D) antiferromagnetic compound $(CD_3)_4NMnCl_3$ (TMMC) in a transverse magnetic field turns out to be a very good realization of sine-Gordon chains below $T \simeq 5$ K.[4] By neutron inelastic-scattering (NIS) experiments[5] and nuclear spin-lattice relaxation-time (T_1) measurements[6] a very accurate description of the soliton regime was possible by analyzing in detail the shape, the intensity, and the width of the staggered mode ($q \simeq \pi$). This mode was identified as the "flipping" mode since its energy (frequency) width gives a direct evaluation of the rate at which the spins of the different sublattices are flipped after each passage of a soliton. One was led to the conclusion that in the very pure crystals investigated so far, the magnetic solitons behave essentially like noninteracting free particles. This model implies that the motion of each soliton remains "coherent" along its trajectory. In other words, it is assumed that the soliton motion is not affected by any collisions and/or any scattering by impurities or defects in the chain. This approximation is valid as long as the soliton mean free path λ remains longer than the inverse soliton density ($n_S\lambda \gg 1$). Otherwise an "incoherent" or diffusive regime is expected for $n_S\lambda \ll 1$.[5]

The experimental results discussed in this Letter were obtained on different crystals of TMMC containing small amounts of magnetic impurities (Cu) with concentrations $c = 0.4\%$, 0.8%, and 1%. For comparison, some measurements were also performed on a pure sample: $c = 0$. In these crystals, the magnetic resonance spectrum of ^{14}N nuclear spins was observed (instead of ^{15}N in an enriched crystal of TMMC used in the work of Boucher and Renard.[6]) In the 1D paramagnetic phase the nuclear spectrum is composed of four well-separated lines for the chosen orientation of the magnetic field perpendicular to the chain axis. Two of these lines result from the superposition of two others (which can be separated by changing the orientation). If we take into account the quadrupolar interaction of ^{14}N nuclear spins, these three pairs of lines can be attributed to the three crystallographic domains which exist below 120 K in TMMC. T_1 measurements have been performed on one of these lines, maintaining always the same experimental conditions, as a function of field and temperature ($20 \lesssim H \lesssim 60$ kOe and $1.4 \lesssim T \lesssim 4$ K). All the T_1 data presented here were obtained in the

1D paramagnetic phase. This point was checked by following the shape of the ^{14}N spectrum: At the 3D ordering transition, a characteristic splitting of the lines occurs. This behavior was observed on both pure and impure samples and was used to distinguish 1D paramagnetic from 3D ordered phases.

Concerning the soliton dynamics to be discussed in the following, we first present briefly an interesting comparison of neutron and T_1 data between the pure sample ($c=0$) and two samples having almost the same concentration of impurities ($c=0.8\%$ for T_1 data and 1% for neutron data). The NIS experiments were performed on the IN12 spectrometer at the Institut Laue-Langevin, with the same conditions as Regnault *et al.*[7] (in particular, the energy resolution was $\Delta\omega \simeq 0.005$ THz). The field dependence of the intensity of the flipping mode at given temperatures as probed (at the wave vector $\vec{Q} = [0.25, 0, 1]$) by NIS measurements is shown in the upper part of Fig. 1. For $c=0$, one observes a sharp maximum that we know results from 3D ordering above the critical field H_c since the emergence of magnetic Bragg peaks was also observed at the same field for the same temperature (for $T=2$ K, $H_c \simeq 40$ kOe). No maximum (and no magnetic Bragg peak) is observed for $c=1\%$, supporting further the NMR result that the spin system remains 1D in the reported field and temperature ranges. The nuclear relaxation T_1^{-1} is shown in the lower part of Fig. 1 for the same field range. For each temperature, a sharp maximum is now observed for both $c=0$ and $c \neq 0$. For $c=0$, the comparison with the neutron data proves that the maximum in T_1^{-1} corresponds to the 3D ordering (the same value of H_c is obtained at the same temperature). This cannot be the case for $c=0.8\%$ since we know both from ^{14}N NMR line shape and neutron data (for $c=1\%$, but the slight difference in c does not change this conclusion) that there is no 3D ordering in the samples with impurities in the field and temperature ranges examined. Rather, we claim that the maximum in T_1^{-1} is a *dynamical* effect in the 1D paramagnetic phase: It corresponds to the condition where the nuclear Larmor frequency ω_N becomes equal to the flipping rate.

In an attempt to analyze our T_1 data, we consider first the coherent soliton model.[8] For this case we have shown[7] that

$$T_1^{-1} \simeq \Gamma/(\omega_N^2 + \Gamma^2), \tag{1}$$

where the flipping rate Γ is given by $\Gamma = 4n_s v_\theta/\sqrt{\pi}$ as a function of the soliton density n_s and the thermal velocity v_θ. It can be written in terms

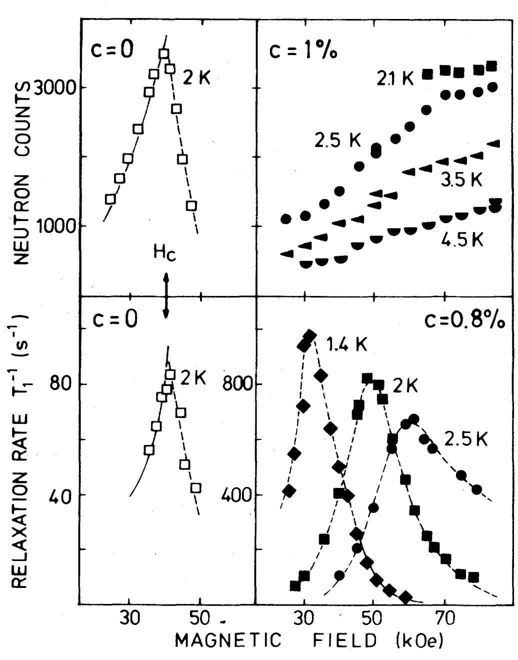

FIG. 1. Intensity of the flipping mode ($q \simeq \pi$ and $\omega \simeq 0$) as observed by neutron inelastic scattering and ^{14}N nuclear relaxation rate T_1^{-1} as a function of magnetic field for pure ($c=0$) and impure ($c \neq 0$) TMMC samples. The full lines are obtained from Refs. 5 and 6 while the dashed lines are aids to the eye.

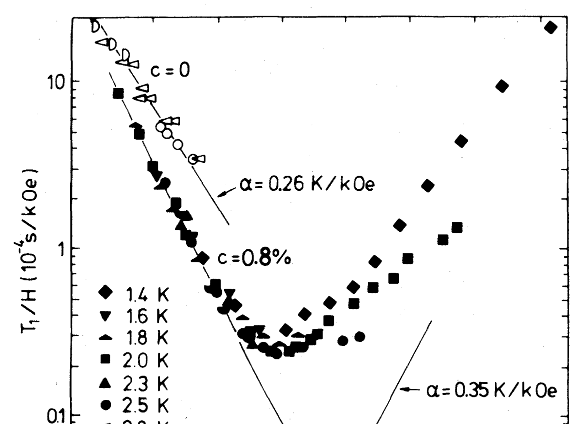

FIG. 2. Experimental values of T_1/H for pure ($c=0$) and impure ($c \neq 0$) TMMC samples compared with the universal behavior in H/T predicted by the coherent model (full lines).

of field and temperature as $\Gamma = BH\exp(-\alpha H/T)$ with $B \simeq (8/S\pi)\alpha \simeq \alpha$. The parameter α gives a direct measure of the soliton energy ($E_s = \alpha H$) and can be determined experimentally. The low-frequency limit, $\omega_N \ll \Gamma$, corresponds to the low-field data for which we expect $T_1/H \sim \exp(-\alpha H/T)$. The curves shown in Fig. 2 are obtained by fitting Eq. (1) to the low-field data. For $c = 0$, we deduce $\alpha \simeq 0.26$ K/kOe in complete agreement with our previous NIS and T_1 measurements on ^{15}N.[6] For $c = 0.8\%$, we obtain $\alpha \simeq 0.35$ K/kOe, and approximately the same value for $c = 0.4\%$ (for clarity the data corresponding to $c = 0.4\%$ are not shown in Fig. 2). Within the coherent model, one is led to the conclusion that the presence of impurities would yield a large increase of the soliton energy (35% larger than for the pure material). To account for this effect we have calculated the corrections which might result from the fact that the chains are finite. We obtain an apparent change in α of only a few percent, which lies within the experimental accuracy. This cannot explain our result. Other arguments against the coherent model are found in the position of the minimum in Fig. 2, which is not explained by the theory, and also in the high-field data. They correspond to the high-frequency limit, $\omega_N \gg \Gamma$, for which Eq. (1) predicts again a universal dependence on H/T according to $T_1/H \sim \exp(+\alpha H/T)$. Instead one observes in Fig. 2 that the points are spread out for large values of H/T.

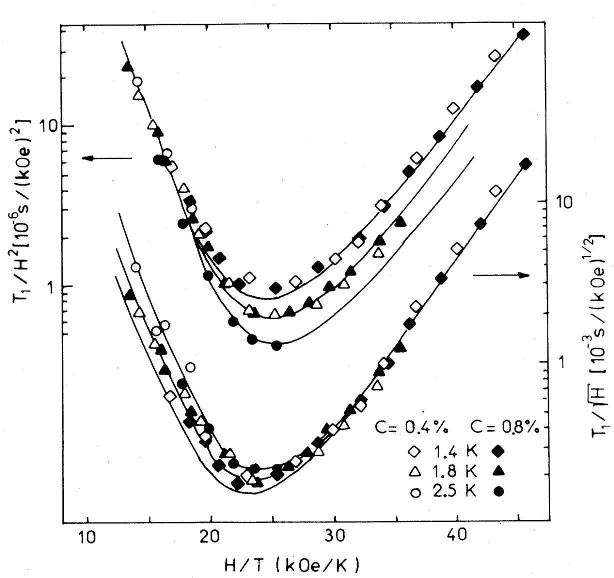

FIG. 3. Experimental values of T_1/H^2 (right-hand side) and $T_1/\sqrt{H}$ (left-hand side) for impure (c = 0.4% and 0.8%) TMMC samples compared with the universal behavior in H/T predicted by the incoherent or diffusive model (full lines).

As a result of scatterings on the magnetic impurities the soliton motion can become incoherent. According to Mikeska,[8] the flipping mode is the spectral representation of $F(t) = \exp[-2N(t)]$ where $N(t)$ counts the number of "uncorrelated" flippings—uncorrelated in the sense given in Ref. 2 for the uncorrelated displacement—from the passages of the solitons at a fixed point coming from both sides. To evaluate $N(t)$ we have followed the analysis of Gunther and Imry.[2] We have assumed that the probability for a particle to be found at a certain distance after a time t is given by the 1D random-walk model. Then, in the long-time limit $t \to \infty$ we obtain $2N(t) = (\Delta t)^{1/2}$ with the flipping rate given by $\Delta = \Gamma^2\tau_D$ where τ_D^{-1} defines the damping rate associated with the diffusion coefficient through $D = v_\theta^2\tau_D$. The Fourier transform of $F(t)$ has been calculated numerically and the resulting expression for T_1 has been fitted to the data, yielding the curves shown in Fig. 3. For $\omega_N \ll \Delta$ and $\omega_N \gg \Delta$ we can use the following asymptotic expressions:

$$T_1^{-1} \sim 2/\Delta \sim (\Gamma^2\tau_D)^{-1}, \tag{2a}$$

$$T_1^{-1} \sim \tfrac{1}{8}(\pi\Delta)^{1/2}/\omega_N^{3/2} \sim \Gamma H^{-3/2}\sqrt{\tau_D}. \tag{2b}$$

In the low-frequency limit $\omega_N \to 0$ a universal dependence on H/T is therefore obtained for $T_1/H^2 \sim \exp[-2\alpha H/T]$ while in the high-frequency limit $\omega \to \infty$, it is for $T_1/\sqrt{H} \sim \exp(+\alpha H/T)$. These limiting behaviors are quite well illustrated in Fig. 3. For the two samples with $c = 0.4\%$ and 0.8% we obtain the same value $\alpha \simeq 0.26$ K/kOe, which is equal to the value deduced previously for the pure sample, $c = 0$ (but within the coherent model[7]). This means that the impurities do not change appreciably the soliton energy, in agreement with our previous calculation. The change observed experimentally results from a different dynamical behavior: In the presence of impurities the magnetic solitons behave diffusively in TMMC. More surprising is that the damping rate coincides for the two impure samples. We obtain $\tau_D^{-1} \simeq 8\times10^{10}$ rad/s. For $H/T = 10$ kOe/K this value of τ_D leads to a mean free path $\lambda = v_\theta\tau_D \sim 100$ lattice units, while $n_s^{-1} \sim 300$. Since $n_s v_\theta(\sim\Gamma)$ decreases exponentially with H/T, we thus check that the condition for a diffusive behavior ($n_s\lambda \ll 1$) is practically fulfilled in the whole experimental range.

As in Ref. 2, we can consider the effect of a finite soliton lifetime τ_l which for $t \gg \tau_D$ leads to $2N(t)=\Delta' t$ with a flipping rate $\Delta' \sim \Gamma(\tau_D/\tau_l)^{1/2}$. In the diffusive regime $(\tau_l \gg \tau_D)$ the corresponding lifetime path is given by $l^2 = v_\theta^2 \tau_D \tau_l$ and one gets $\Delta' \sim \Delta/n_s l$. If $n_s l \gg 1$ as claimed in Ref. 2, the effect of the finite lifetime on the flipping mode is negligible. Finally we can assume the solitons to be reflected at the ends of the chains as antisolitons. Unlike the case considered by Gunther and Imry,[2] we do not expect any qualitative change in the dynamics since, for a given particle, $N(t)$ is proportional to the number of passages at a given point and not to the square of this number.

The purpose of this Letter was to display the drastic changes observed in the soliton dynamics by the presence of a small amount of magnetic Cu impurities in the chains of TMMC. The diffusive model gives a very good interpretation of the data. We have not considered the microscopic origin of the diffusion and the damping rate τ_D^{-1} remains a phenomenological parameter. In TMMC, all the relevant quantities are known with accuracy, in particular the Mn-Cu coupling. A quantitative analysis should be possible, from which decisive progress in the study of the diffusive regime for a soliton gas can be expected.

We hope that these new results will stimulate more experimental and theoretical works.

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Self-Consistent Nonequilibrium Description of Phase-Slip States in Superconducting Filaments

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Inhomogeneous dissipative states found in a current-driven superconducting filament have been investigated with explicit allowance for depairing and coupling between pairs and quasiparticles off equilibrium. The resulting charge imbalance is insensitive to the gapless approximation used over a wide range of parameters; it can induce additional synchronized phase-slip centers, and leads to novel effects related to a mismatch between the core and the wings of each phase-slip center if local equilibrium cannot be maintained.

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Slightly below the transition temperature T_c, the dc characteristic of a long inhomogeneous superconductor with small transverse dimensions driven by a direct current I exhibits a remarkable sequence of overlapping branches with time-average voltages $\bar{V}_m$. In the mth state, $d\bar{V}_m/dI$ is effectively the resistance of m normal segments with resistivity ρ_N and length $2\Lambda_V$ in series. Each segment can be associated with a localized phase-slip center (PSC)[1] because the increase in the phase difference φ implied by Josephson's relation $\hbar\dot{\varphi}=2eV$ must be compensated by -2π jumps in the phase of the order parameter $\Delta e^{i\chi}$.[2] The accompanying periodic collapse of Δ presumably heals over a few coherence lengths ξ, whereas the electrochemical potential μ decays outside the core of each PSC over a distance $\sim\Lambda_V$ related to the diffusion length Λ_0 of excited quasiparticles.[1]

Except in strong-coupling superconductors or

