Energy Absorption from an Oscillating Magnetic Driving Field by Soliton Motions in the Quasi One-Dimensional Ferromagnet $[(CH_3)_4N]NiCl_3$ (TMNC)

L. J. de Jongh, C. A. M. Mulder, R. M. Cornelisse, and A. J. van Duyneveldt
Kamerlingh Onnes Laboratory, University of Leiden, Leiden, The Netherlands

and

J. P. Renard
Institut d'Electronique Fondamentale, Université Paris XI, F-91405 Orsay, France

(Received 1 June 1981)

Experimental evidence for one-dimensional domain-wall relaxation above T_c in the quasi one-dimensional ferromagnet $[(CH_3)_4N]NiCl_3$ has been obtained from ac susceptibility measurements in the frequency range 10 Hz $< \nu <$ 30 MHz. The field and temperature dependence of the absorption is well explained by the interaction of π solitons with the oscillating magnetic field.

PACS numbers: 75.50.Dd, 75.10.Hk, 75.60.Ch

New experimental methods to study solitary excitations are highly desirable in view of the recent vivid interest in nonlinear dynamics.[1] Lately, it has been predicted[2] that solitons may occur in magnetic chains, where they correspond to domain walls ("kinks") separating, e.g., the spin-up and spin-down regions in a ferromagnetic chain, or similarly the two degenerate ordered configurations of an antiferromagnetic chain. That the motion of a Bloch wall in a ferromagnetic chain is described by the sine-Gordon equation was in fact already shown much earlier by Enz.[3] Such solitons are thermally excited, and their existence in experimental quasi one-dimensional (1D) magnetic systems has already been probed by neutron scattering,[4] NMR nuclear spin-lattice relaxation,[5] and Mössbauer linewidth measurements.[6]

In this note we describe an extremely simple and direct way of coupling to solitons in a ferromagnetic chain, namely by their interaction with an applied oscillating magnetic field. Experimental data are presented on the frequency-dependent ac susceptibility of the quasi 1D ferromagnet $[(CH_3)_4N]NiCl_3$ (TMNC), which clearly evidence 1D domain-wall relaxation in the *paramagnetic* region. Both the temperature and the field dependences of this relaxation process are well explained on the basis of the soliton model.

Previous susceptibility[7,8] and specific-heat[9] studies on TMNC have shown that the compound is composed of ferromagnetic chains running parallel to the hexagonal c axis, with *intrachain* exchange $J/k_B \simeq 1.7$ K. A strong crystal-field anisotropy of the XY type, $D^{XY}/k_B \simeq 3.4$ K, forces the magnetic moments to lie within the (basal) plane perpendicular to c. The susceptibility $\chi_{\perp c}$ shows a sharp peak at the transition temperature $T_c = 1.195$ K, below which the chains become 3D ordered in an antiparallel way because of the weak *interchain* interaction $J'/k_B \simeq -0.015$ K. The latter value is derived[7] from the saturation field $H_c = 2400$ Oe at $T = 0.4$ K. A weak Ising-type anisotropy, $D^I/k_B \simeq 0.01$ K, establishes a preferential direction within this easy plane. The value of D^I follows from the spin-flip field $H_{sf} \simeq 400$ Oe at $T = 0.4$ K.[7] The main source of D^I is thought to be dipolar anisotropy combined with a slight monoclinic distortion occurring at high temperature. The resulting broken symmetry within the basal plane leads to a three-domain structure, with the directions of the moments related by 120° rotations. From the values of D^{XY} and D^I it follows that for $T \gg T_c$ the system will behave as a 1D XY ferromagnet, for which the correlation length is $l \propto T^{-1}$, whereas for $T \to T_c$ a crossover to Ising behavior will occur, leading to $l \propto \exp[2JS(S+1)/k_BT]$.[10] From the observed[7] temperature dependence of $\chi_{\perp c}$ we may deduce that the crossover occurs near $T \simeq 2$ K.

The complex ac susceptibility, $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, of a single crystal was measured as a function of frequency $\nu = \omega/2\pi$ in the range 10 Hz–1 MHz in two different apparatus.[11] (A few data for $\nu > 10$ MHz were kindly provided by the group of Dr. J. C. Verstelle.) Since the absorption χ'' had to be measured with great precision, the correct phase setting was checked prior to each measurement by replacing the TMNC sample by a paramagnetic Mn^{2+} salt, and adjusting the phase in zero field. This precaution proved to be crucial since $\chi' \gg \chi''$ for $T \to T_c$. The amplitude h_0 of the ac field $h = h_0 e^{i\omega t}$ was typically about 1 Oe. Parallel to h a constant field H_0 could be applied.

Representative data for χ' and χ'' are shown as a function of ν (at $H_0 = 665$ Oe) and of H_0 (at $\nu = 10.6$ kHz) in Figs. 1 and 2, respectively, for a number of temperatures. The applied field $H = H_0 + h_0 e^{i\omega t}$ is within the basal plane (in which χ is isotropic because of the three-domain structure). In Fig. 1 three different and, fortunately enough, well-separated relaxation processes can be discerned. For $\nu > 10$ MHz, one just observes the onset of spin-spin relaxation, to be expected in this higher-frequency range. For $\nu < 500$ Hz, the tail of the spin-lattice relaxation process can clearly be seen. The latter assignment is confirmed by the fact that this absorption vanishes for $H_0 \to 0$. (A paramagnet cannot display spin-lattice relaxation in zero field.) Additional data at 4.2 K in fields up to $H_0 = 24$ kOe also indicate that the spin-lattice relaxation at $T \simeq T_c$ should occur for $\nu < 1$ kHz.

On the other hand, the relaxation process observed in the intermediate range, 500 Hz $< \nu < 5$ MHz, displays a quite novel behavior as a function of H_0 and T, and is ascribed to soliton motions. In fact, this frequency range corresponds quite well with those met in previous experiments on domain-wall relaxation,[12,13] the important difference being that those experiments dealt with 3D ordered ferromagnetic materials, whereas here we are seeing domain-wall relaxation inside 1D correlated chain segments in the *paramagnetic* phase. Since $\chi'' \neq 0$ for $H_0 \to 0$ (cf. Fig. 2), this absorption cannot be a spin-lattice process. However, it agrees with the soliton model, since the presence of D^{I} makes possible the excitation of solitons even when $H_0 = 0$. For $T \to T_c$, $\chi''(H=0)$ increases at first, but vanishes rapidly below T_c in agreement with the fact that the establishment of 3D magnetic order will gradually block the soliton motions. We recall that the ordering between the chains is *antiferromagnetic*, so that the 1D ferromagnetic domains cannot combine to form 3D ones below T_c.

As long as D^{I} is the predominant factor that breaks the symmetry in the basal (XY) plane, the solitons may correspond to rotations over an angle π or 2π of the spins (cf. the inset in Fig. 1). However, if H_0 becomes sufficiently strong to align the moments, the π solitons will be suppressed. This explains the strong field dependence of χ'' in Fig. 2, because only π solitons will be effective for the relaxation process, as

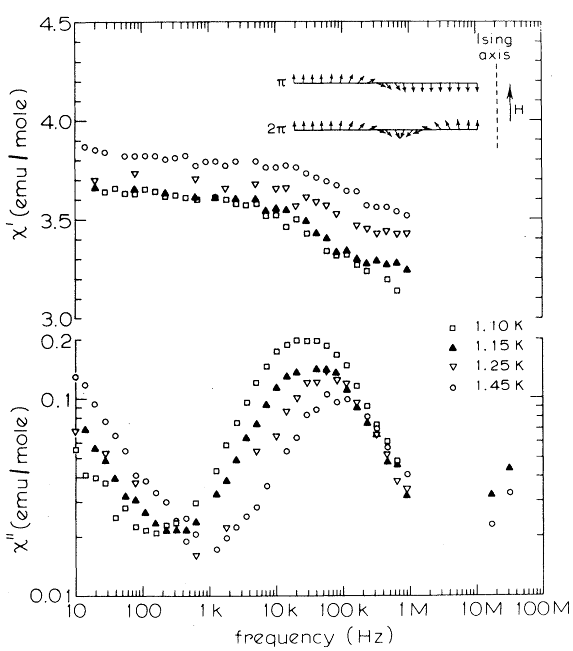

FIG. 1. Dependence on frequency ν (log scale) of the real part χ' (linear scale) and the imaginary part χ'' (log scale) of the ac susceptibility of TMNC. Inset shows the π and 2π solitons occurring in the Ising-type ferromagnetic chain.

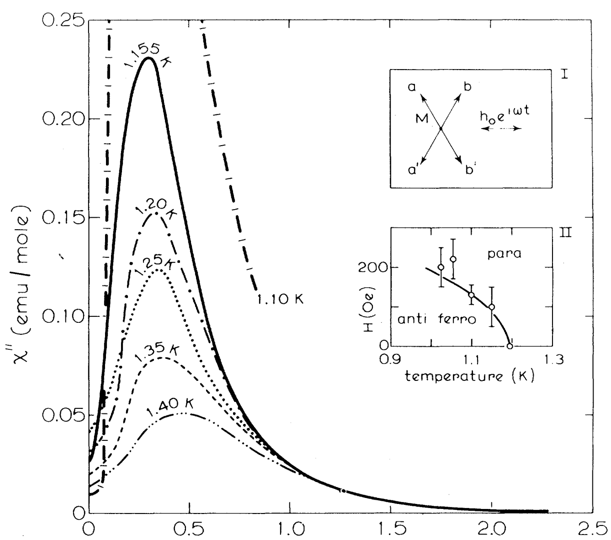

FIG. 2. Field dependence of the absorption χ'' at $\nu = 10.6$ kHz for different temperatures (the curve for $T = 1.10$ K reaches a maximum value $\chi'' \simeq 0.35$ emu/mole at $H_0 \simeq 250$ Oe). Inset I illustrates the relaxation process, whereas inset II denotes the antiferromagnetic phase diagram deduced from the maxima in the χ' vs H isotherms.

is immediately obvious by considering that the field H in the inset in Fig. 1 may only yield a displacement of the π kink, whereas the 2π kink will not move (only its width will vary with H). The relaxation may also be visualized as shown in inset I in Fig. 2. If h is at some angle with the spin direction in the easy plane, it will cause an oscillation of the (correlated ferromagnetic) moments around this axis. In the absence of relaxation the magnetization M will move in phase with h in between say a and b, or equivalently a' and b'. That is, if h points to the left, the in-phase position corresponds to a or a'. A passing π soliton will reverse M (e.g., from a to b'), resulting in the phase angle φ and thus in an absorption χ'' $\propto \sin\varphi$. On the other hand, a passing 2π soliton will leave M eventually in its original position.

In inset I in Fig. 2 the direction of h is perpendicular to the easy axis, leading to maximum phase angle φ. However, as mentioned above, for $H_0 \rightarrow 0$ a three-domain structure is formed, with the easy axes of different domains at angles of 120°. Therefore, for $H_0 \ll H_{sf}$ the majority of the moments will be at some small angle with H_0, and only for $H_0 \simeq H_{sf}$ will the spins be all nearly perpendicular to H_0 and the relaxation be a maximum. In Fig. 2 the χ'' maxima are found at 300–500 Oe, which is indeed close to the value of H_{sf} $\simeq 400$ Oe. For larger fields H_0, the moments will become aligned so that $\chi'' \rightarrow 0$ by the suppression of the π solitons. For $T < T_c$ and $H_0 \rightarrow 0$ the ordered region is entered and the solitons become immobile. The magnetic phase diagram, deduced from the observed anomalies in χ' as a function of H_0, is given in inset II in Fig. 2 and at 1.10 K (cf. Fig. 2) the maximum in χ' is found at $H_0 \simeq 130$ Oe only. We mention that for 1 kHz $< \nu <$ 1 MHz the field dependence of χ'' remains qualitatively quite similar, and also that the domain relaxation process is *not* observed with $H \parallel c$ axis, in agreement with the above-outlined soliton relaxation model.

The strong temperature dependence of χ'' can be qualitatively explained as follows. One may expect $\chi'' \propto n_s l$, where n_s is the soliton density. In the Ising region below 2 K one has $l \propto \exp[2JS(S+1)/k_BT]$. For the ferromagnetic chain with classical spins and Ising-type anisotropy it follows from Mikeska's work[2] that $n_s = (2D^{\mathrm{I}}/\pi J)^{1/2}(E_0/k_BT)^{1/2}\exp(-E_0/k_BT)$, where $E_0 = S^2(16DJ)^{1/2}$ is the soliton rest energy. For TMNC ($S=1$) one calculates $E_0/k_B = 0.5$ K. This falls short of the experimental[7] Ising crossover temperature T^{I} $\cong 2$ K, although at first thought the two should be nearly the same. However, quantitative agreement should not be expected, e.g., since both Mikeska's expression for E_0, as well as the argument[14] leading to $k_BT^{\mathrm{I}} \propto S(DJ)^{1/2}$, is based upon the model of simple uniaxial Ising anisotropy, whereas experimentally one has an orthorhombic anisotropy with $|D^{XY}/D^I| \gg 1$. In that case a large enhancement of the experimental T^{I} with respect to the above expression is to be expected.[15] Further enhancement will result from the weak but finite interchain couplings.[13] Lastly, we remark that Mikeska's theory applies to classical spins in the continuum approximation, whereas in TMNC one has quantum mechanical spins on discrete lattice sites.

Although quantitative agreement will thus be poor, the above argument predicts $\chi'' \propto \exp(-\Delta/k_BT)$, with a Δ of a few degrees Kelvin. Indeed the temperature dependences of the χ'' maxima in Figs. 1 and 2 can be fit by an exponential with an energy of about 6 ± 3 K, i.e., of the right order of magnitude. Furthermore, with increasing ω the wall displacements due to $h_0 e^{i\omega t}$ will become small compared to l, explaining why χ'' is nearly independent of temperature for 100 kHz $< \nu <$ 1 MHz in Fig. 1.

We end by invoking the celebrated model for domain-wall motion already used by Kittel and Galt,[12] in which the moving wall is treated mathematically as a point mass in classical mechanics, performing a linear motion under the influence of a time-varying driving force:

$$m\ddot{z} + \beta\dot{z} + \alpha z = 2Mh_0 e^{i\omega t}. \qquad (1)$$

Although originally meant to describe the motion along an axis (z) of a planar wall in a 3D ferromagnet, application to our present problem appears obvious. We recall[1] that the soliton may indeed be viewed as a moving particle of a certain mass m. The effect of external and damping forces appears to have also been considered already.[1] Although the frictional and pinning forces $\beta\dot{z}$ and αz experienced by a magnetic soliton in its motion along the chain may be introduced in a pure phenomenological sense, we consider as likely sources the presence of the *interchain* interactions, which, even in the paramagnetic region, will hamper the free motions of the solitons. Lattice defects and impurities will also play a role, the more so if some degree of correlation between the soliton motions in adjacent chains would be established through the *interchain* interactions. In this respect we note that the exchange field associated with the *interchain* interactions amounts to 1200 Oe at $T = 0$ K, whereas $h_0 \simeq 1$ Oe.

If then Eq. (1) is accepted, the desired relaxation behavior will be obtained in the limit $\beta^2 \gg m\alpha$, in which case the susceptibility is given by the Debije formula[12,13]:

$$\chi/\chi_0 = (\chi' - i\chi'')/\chi_0 = (1+\omega^2\tau^2)^{-1} - i\omega\tau(1+\omega^2\tau^2)^{-1},$$

where the relaxation time $\tau = \beta/\alpha$, and $\chi_0 = 4M^2/\alpha d$ with d the average distance between the walls. Experimentally,[12,13] deviations from this behavior are observed in that the χ'' vs ω curves are often flattened, which is then ascribed to a distribution of relaxation times. We likewise observe such flattening with increasing T and H_0. Only for $T \simeq T_c$ and $H_0 \simeq 500$ Oe does the $\chi''(\omega)$ curve approximate the Debije form. Since there will certainly be a distribution in the lengths of the correlated chain segments, and since these correlated 1D regions will be randomly distributed in adjacent chains, it follows that the effective *interchain* interactions will not be uniform along the chain. If the latter are indeed responsible for the αz and $\beta \dot{z}$ terms this will lead to a distribution in τ, and the broadening will be enhanced if l is decreased, e.g., by increasing T.

Values for the parameters m, β, and α may be estimated as follows. For m one has the formula[12,13] $m = (2\pi\gamma^2\Delta)^{-1}$, where $\gamma \simeq 1.8\times 10^7$ Oe^{-1} s^{-1} is the gyromagnetic ratio and the domain-wall width $\Delta = a_0(J/D^{\mathrm{I}})^{1/2} \simeq 4\times 10^{-7}$ cm, leading to $m \simeq 1\times 10^{-9}$ Oe^2 s^2 cm^{-1}. The formula for the soliton density n_s in the above leads to the estimate $d \simeq 10^{-6}$ cm at $T \simeq 1.3$ K. Assuming that the 1D correlated regions contain about 10% of the spins, the value $M \simeq 10$ G is obtained for the magnetization. We may then estimate α from $\chi_0 = 4M^2/\alpha d$ by taking $\chi_0 \simeq \chi'(\nu = 1\text{ kHz}) - \chi'(\nu = 1\text{ MHz}) \simeq 0.5$ cm^3/mole, obtaining $\alpha \simeq 1\times 10^{11}$ Oe G cm^{-1}. From $\tau = \beta/\alpha$ and the average relaxation time of $\tau \simeq 3\times 10^{-5}$ s, we find $\beta \simeq 3\times 10^6$ Oe G s cm^{-1}. One easily verifies that the condition for relaxation $\beta^2 \gg m\alpha$ is certainly fulfilled.

We would like to thank N. A. H. Verhaar for supplying the data at $\nu > 10$ MHz, and H. W. Capel, L. J. de Haas, and J. C. Verstelle for stimulating discussions.

[1]See, e.g., A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Physica (Utrecht) 1D, 1 (1980).

[2]H. J. Mikeska, J. Phys. C 11, L29 (1978), and 13, 2913 (1980).

[3]U. Enz, Helv. Phys. Acta 37, 245 (1964), and Phys. Rev. 131, 1392 (1963).

[4]J. K. Kjems and M. Steiner, Phys. Rev. Lett. 41, 1137 (1978); J. P. Boucher, L. P. Regnault, J. Rossat-Mignod, J. P. Renard, J. Bouillot, and W. G. Stirling, Solid State Commun. 33, 171 (1980).

[5]J. P. Boucher and J. P. Renard, Phys. Rev. Lett. 45, 486 (1980).

[6]R. C. Thiel, H. de Graaf, and L. J. de Jongh, Phys. Rev. Lett. 47, 1415 (1981).

[7]C. Dupas and J. P. Renard, J. Chem. Phys. 61, 3871 (1974), and J. Phys. C 10, 5057 (1977).

[8]J. V. Lebesque and P. van Meeuwen, Commun. Phys. 2, 131 (1977).

[9]K. Kopinga, T. de Neef, W. J. M. de Jonge, and B. C. Gerstein, Phys. Rev. B 13, 3953 (1976).

[10]J. P. Boucher, L. P. Regnault, J. Rossat-Mignod, and J. Villain, J. Magn. Magn. Mater. 14, 155 (1979).

[11]H. A. Groenendijk, A. J. van Duyneveldt, and R. D. Willett, Physica (Utrecht) 101B, 320 (1980); A. J. de Vries and J. W. M. Livius, Appl. Sci. Res. 17, 31 (1967).

[12]Ferromagnetic domain theory is treated by C. Kittel and J. K. Galt, Solid State Phys. 3, 437 (1956). Recent work on wall dynamics is reviewed by F. H. de Leeuw, R. van den Doel, and U. Enz, Rep. Prog. Phys. 43, 689 (1980).

[13]Experimental examples may be found in, e.g., E. Kneller, in *Ferromagnetismus* (Springer-Verlag, Berlin, 1962), and J. Smit and H. P. J. Wijn, in *Ferrites* (Philips' Technical Library, Eindhoven, 1959).

[14]One may put $k_B T^{\mathrm{I}} = D^{\mathrm{I}} l$. With (Ref. 10) $l = 4JS^2/k_B T$ (for 1D Heisenberg model) the required result follows.

[15]F. Boersma, W. J. M. de Jonge, and K. Kopinga, Phys. Rev. B 23, 186 (1981).