Evidence for Soliton Modes in the One-Dimensional Ferromagnet $CsNiF_3$

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Evidence for solitons moving along the ferromagnetic chains in $CsNiF_3$ has been obtained by inelastic neutron scattering. As predicted by Mikeska the scattering is found at low q, around zero energy. The soliton activation energy, $8m$, is determined via the temperature and field dependence of the intensities (m is the effective mass of the quasiparticle soliton). At $H=5$ kG we find $8m/k_B=27$ K in reasonable agreement with the predicted value, as is the energy width at $q=0.1$.

Considerable interest has recently been focused on nonlinear effects in the excitation spectrum of solids.[1-5] One-dimensional (1D) model systems have been given special attention since analytic solutions exist to the nonlinear equations of motion like the sine-Gordon equation. Most well known are the soliton solutions.

Mikeska recently showed[6] that the spin dynamics of a 1D ferromagnet with easy-plane anisotropy like $CsNiF_3$ in an applied symmetry-breaking field is also governed by the sine-Gordon equation. He developed a classical continuum theory which documents the analogy between equations of motion for this spin Hamiltonian and those pertinent to the classical sine-Gordon system: namely, the array of mechanically coupled restricted pendula. In this analogy the magnetic field corresponds to the gravitational field and the exchange to the spring coupling constant.

The thermodynamics of such 1D sine-Gordon systems has been discussed by several authors.[2-5] A simple and appealing physical picture has emerged in which the solitons can be regarded as quasiparticles with a finite mass or activation energy and a thermal distribution of velocities. Most of the discussions have dealt with problems related to structural transitions and charge density waves, but so far only indirect experimental evidence is available to sustain the soliton picture of the dynamics.

In this Letter we report observations via inelastic neutron scattering of solitons moving along the chains in $CsNiF_3$. Our data are compared to the predictions made by Mikeska based on the well-known microscopic Hamiltonian for for $CsNiF_3$.

The system we studied, $CsNiF_3$, has hexagonal crystal structure, $P6\ 3/mmc$, with lattice constants $a=b=6.21$ Å and $c=5.2$ Å. The structure consists of NiF_6 octahedra sharing common faces along the $\vec{c}$ axis, well separated by Cs ions. Hence the exchange coupling of the Ni^{2+} ions is much stronger along $\vec{c}$ than along $\vec{a}$ and $\vec{b}$. The results of the different studies on $CsNiF_3$ have been reviewed by Steiner, Villain, and Windsor.[7] The conclusion is that the system consists of ferromagnetic chains running along $\vec{c}$ with strong coupling within the chains and without significant coupling between chains at temperatures above 3 K, T_N being 2.7 K. The magnetic moments are bound to the ab plane by a strong single-site anisotropy. In this plane they can rotate freely. It was found that the following Hamiltonian is appropriate for $CsNiF_3$ at $T>3$ K:

$$\mathcal{H} = -2J\sum_i \vec{S}_i\cdot\vec{S}_{i+1} + A\sum_i (S_i^z)^2 - g\mu_B H^x \sum_i S_i^x \quad (1)$$

with $J/k=11.8$ K; $A/k=9.5$ K.

The present experiments were carried out on a cold-neutron source, triple-axis spectrometer at the DR3 reactor at Risø. The incident energy was fixed at 5 meV and a cooled Be filter was used to to remove higher-order contamination. The horizontal collimators were 60′, 45′, 52′, and 60′, from reactor to detector, and pyrolytic graphite crystals were used as monochromator and analyzer. The energy resolution, as measured by the full width at half-maximum of the incoherent elastic peak from the sample, was 0.18 meV. The $Cs^{58}NiF_3$ sample was a good-quality single crystal from Crystal Tec, Grenoble, with a volume of 1 cm^3. In order to minimize the incoherent scattering the sample only contained the ^{58}Ni isotope. It was mounted in a split-coil superconducting magnet with vertical field. The horizontal scattering plane contained the ac crystal plane. The measurements were made in the (002) zone and especially the inelastic spectrum at (0, 0, 1.9) was studied in detail as a function of temperature and field in the ranges $0<T<15$ K and $0<H<36$ kG. The main contribution to the peak at zero energy transfer comes from elastic incoherent scattering, and the following procedure was used to measure and subtract this background component. We assume that at high field and low temperature the elastic peak is solely due to the incoherent scattering and thus the observed spectrum at 3.1 K and 30 kG, which is shown as the full line in the upper part of Fig. 1, was subtracted from all the other spectra. The resulting background-corrected spectrum is shown in the lower part of Fig. 1 for the data presented in the upper part. This spectrum clearly has three components: two relatively narrow peaks at ± 0.55 meV corresponding to the creation and annihilation of spin waves propagating along the chains, and a broader quasielastic distribution whose energy width is considerably larger than the experimental resolution. In order to get a quantitative description of this feature, the observed distributions were least-squares fitted by a calculated line profile which assumed a Gaussian distribution for the central component and Lorentzian profiles for the spin-wave resonances, which have been found in the previous studies of $CsNiF_3$.[7]

The final line profile was obtained by a one-dimensional convolution with a Gaussian resolution function including detailed balance and other trivial experimental factors. The full line in the lower part of Fig. 1 is the result of such line-shape analysis. Fits of a similar quality were obtained for all of the observed spectra and the resulting values for the integrated intensity and for the half-width at half-maximum (HWHM) of the central Gaussian component are summarized in Fig. 2 for a fixed field of 5 kG at different temperatures. The errors that are indicated in this figure are those resulting from the statistical uncertainties and they do not include any estimates of possible systematic errors. The dramatic increase with temperature of the central component of the spectra is accompanied by an almost linear drop in the spin-wave intensity by a factor of 4 in going from 3.1 to 14 K, whereas the spin-wave linewidth does not change significantly and remains small compared to the experimental resolution. The spectrum at (0, 0, 1.9) was also followed as a function of field at 9.5 K. The results for the integrated quasielastic intensity at 9.5 K is plotted in Fig. 3(b). The linewidth in the field scans was found to decrease slowly to about 0.12 meV (HWHM) at 13.2 kG. Scans at

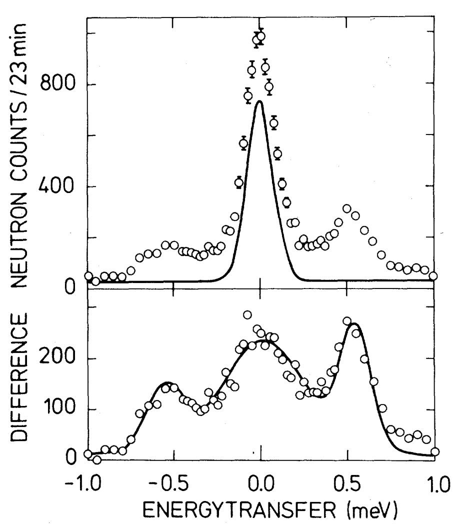

FIG. 1. Upper: Observed inelastic spectrum at (0, 0, 1.9) at $T = 9.3$ K and $H = 5$ kG (circles). The full line is the observed profile at $T = 3.1$ K and 30 kG which is assumed to be the background. Lower: Difference between the two spectra in the upper part of the figure. The full line is the result of a least-squares fit as described in the text.

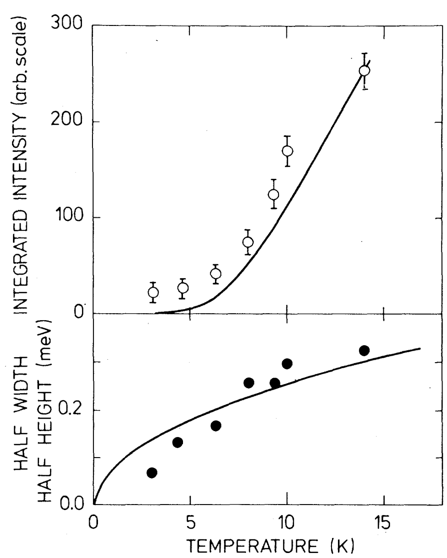

FIG. 2. Temperature dependence of the integrated intensity (upper) and energy half-width at half-maximum (lower) of the quasielastic component at $H = 5$ kG and $q = (0, 0, 1.0)$. The values are derived from the least-squares fitting procedure described in the text. The full lines are the results predicted by Mikeska with no adjustable parameter for the energy width and only a scale factor for the intensity.

different wave vectors were also performed at 5 kG and $T=9.3$ K. These spectra showed a steep increase in the central-component intensity for decreasing q_c, with the spin-wave peaks appearing only as shoulders. No quantitative analysis was attempted on these data.

In order to discuss the experimental data, we compare the quasielastic component to the predictions made by Mikeska[6] for the dynamic structure factor $S_{\rm sol}(\vec{q},\omega)$ in the context of his classical continuum soliton theory,

$$S_{\rm sol}(q,\omega)\propto \frac{\beta e^{-8m\beta}}{cq}\exp\left(-\frac{4\beta m\omega^2}{c^2q^2}\right)\left(\frac{\pi q/2m}{\sinh(\pi q/2m)}\right)^2, \tag{2}$$

where $\beta=(k_{\rm B}T)^{-1}$ and all energies are measured in units of JS. The effective mass m is given by $m=(g\mu_{\rm B}H/2J)^{1/2}$ and the velocity c by $c=S(4AJ)^{1/2}$; $S=1$, $g=2.4$, and $J/k_{\rm B}=11.8$ K. In this classical theory the renormalized value for the anisotropy energy is used,[8] $A/k_{\rm B}=4.5$ K. The solitons considered here have an activation energy of $8m$. This cross section is formally very similar to the scattering from a dilute gas of finite-size particles with mass $8m/c^2$ and a thermal velocity distribution. It contains no adjustable parameter besides a scale factor for the intensity since all constants of the system are well known. In particular, we find immediately for fixed q and H the following properties: The soliton intensity times $\sqrt{T}$ is exponential in temperature with an activation energy of $8m$; the soliton energy width shows a square-root-dependence on temperature with no adjustable parameter. In addition significant soliton scattering should be seen only for small $\vec{q}$. This is because of the spatial dimensions of the solitons considered here: They are 2π solitons, meaning that within one soliton the spin rotates over 2π in the ab plane. The length of the soliton is determined by $1/m$ and reflects the competition between Zeeman energy and exchange energy. The creation of such a soliton involves large spin changes which inhibit a direct excitation of solitons in an inelastic neutron-scattering experiment. Hence the experiment is done on a system with thermally excited solitons, which move along the chains with a certain velocity $u<c$ like a gas of noninteracting particles. The scattering process is therefore similar to the scattering of neutrons from a real gas.

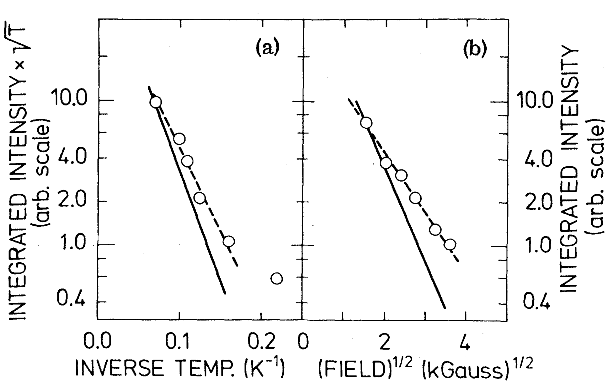

FIG. 3. Semilog plots of the integrated quasielastic intensity at (0, 0, 1.9) vs inverse temperature at $H=5$ kG and (b) vs the square root of the field at $T=9.5$ K. The solid lines are calculated from the Mikeska theory.

When calculating the temperature dependence of the intensity of the soliton peak at fixed $\vec{q}=0.1$ reciprocal lattice units (r.l.u.) and fixed field 5 kG we need to scale the intensities in order to compare with the measured results in Fig. 2. Fixing the scale at 14 K one finds the solid line given in the upper part of Fig. 2. It is evident that the experiment shows the expected increase with temperature for $T>4.0$ K, whereas at the lowest temperatures a signal remains which does not follow the theory. The energy width of the soliton peak can be calculated from Eq. (2) directly without any adjustable parameters. The solid line in the lower part of Fig. 2 is the calculation for $\vec{q}=0.1$ r.l.u. and $H=5$ kG with the above-mentioned constants. Here the agreement with theory is very good except again at the lowest temperatures $T<4.6$ K. We do not have any explanation for the scattering at low temperature, which is also found at zero field. Since the correction procedure involves the subtraction of a relatively large incoherent peak, we cannot exclude a systematic error in all our intensity data which could be of the order of the intensities found at $T<4.6$ K. Such a systematic error would shift the intensity-versus-temperature curve in Fig. 2 as a whole but would not change the experimental increase with temperature; hence it could give an even better agreement between the theory and the experiment. The strong q dependence and the decreasing spin-wave intensities rule out the possibility of a contribution of multiple spin-wave excitations. In what follows we concentrate on the data for $T>6.3$ K. In order to determine the experimental value for the activation energy we plot $\ln(I\sqrt{T})$ versus $1/T$ as shown in Fig. 3(a) where the solid line corresponds to the solid line in the upper part of Fig. 2. The best straight line through the experimental points for $T > 6.3$ K yields [broken line in Fig. 3(a)]

$$8m_{\rm expt} = 27 \text{ K},$$

which compares well with the theoretical results,

$$8m_{\rm theor} = 34 \text{ K},$$

for $H = 5$ kG and $\vec{q} = 0.1$ r.l.u. Following Eq. (2) we see that the H dependence of the soliton intensity is also given by a nearly exponential function for $T = 9.5$ K, namely, $I \propto \exp(-C\sqrt{H})$. We thus plotted in Fig. 3(b) $\ln(I)$ versus $\sqrt{H}$ (H in kG). The solid line is calculated using Eq. (2) and it gives the value $C_{\rm theor} = 1.52$. It is scaled to the experiment at the lowest field. The broken line in Fig. 3(b) is the best fit to our data and yields $C_{\rm expt} = 1$.

The results discussed above can be summarized as follows. We were able to describe our experimental results for the soliton peak around zero energy in the frame of the following picture. The neutrons are scattered from a gas of noninteracting soliton quasiparticles which are thermally excited. The corresponding Mikeska classical theory describes quantitatively the results for fixed field, $H = 5$ kG, and different temperatures, whereas the agreement for fixed temperature, $T = 9.5$ K, and different fields is only qualitative. Especially, the intensity does not fall off with increasing field as rapidly as predicted. Our measured $\vec{q}$ dependence indicates a more rapid variation than predicted theoretically. Despite the lack of direct excitation solitons by means of inelastic neutron scattering our results give direct experimental evidence that the soliton picture is appropriate for the dynamics of the 1D ferromagnet $CsNiF_3$.

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On page 328, Table I, column one, the first value of r_s was incorrectly printed as 1.7. The correct value is 0.7.

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The present address of the author was inadvertantly omitted. It should read as follows:

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