Evidence for solitons in a quantum ($S=\frac{1}{2}$) XY system

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The heat capacity and nuclear spin-lattice relaxation of the quasi-one-dimensional compound $(C_6H_{11}NH_3)CuBr_3$ have been measured for 1.5 K $< T <$ 7 K and magnetic fields up to 7 kG. All data are in good agreement with classical soliton theory, both for in-plane and inclined fields, yielding the first evidence of solitons in a ferromagnetic quantum XY chain.

The suggestion that solitons might be considered as elementary excitations in one-dimensional systems[1] has prompted a large theoretical effort. It appeared that—*in the continuum limit*—the equation of motion of a *classical* ferromagnetic chain with XY anisotropy could be mapped to a sine-Gordon (sG) equation, which is known to support soliton solutions. Next, such a mapping was obtained[2] for several other types of magnetic systems.

The first experimental evidence of solitons in magnetic systems was observed[3] in the neutron scattering cross section of the $S=1$ XY ferromagnet $CsNiF_3$. Allthough this evidence has been disputed,[4,5] more detailed results[6] as well as spin-lattice relaxation time[7] and specific-heat measurements[8] confirmed the relevance of solitons in this system. Recently a soliton contribution was also observed[9,10] in the $S=\frac{5}{2}$ antiferromagnet $(CH_3)_4NMnCl_3$ (TMMC). In both compounds a substantial reduction of the classical soliton rest energy had to be assumed to describe the experimental data. This fact was associated with quantum effects,[11] since the mapping to a sG equation explicitly exploits the classical nature of the spins.

In this Rapid Communication we present the first experimental evidence of solitons in a *real* ferromagnetic quantum ($S=\frac{1}{2}$) chain with XY anisotropy. We measured both the nuclear spin-lattice relaxation (NSLR) time T_1 and the specific heat C of the recently discovered system $(C_6H_{11}NH_3)CuBr_3$. We will show that both the dynamic behavior measured by T_1 and the equilibrium properties monitored by C can be understood if we assume that solitons are the dominant excitations in the appropriate temperature and field range.

$(C_6H_{11}NH_3)CuBr_3$ (CHAB) is the Br isomorph of the Cl compound known as an excellent one-dimensional ferromagnet.[12] Several investigations[13,14] showed that the system can be characterized by the Hamiltonian

$$\mathscr{H}=-2J\sum_i[\vec{S}_i\cdot\vec{S}_{i+1}+(J_{zz}-J)/JS_i^zS_{i+1}^z]-\sum_i g\mu_B\vec{B}\cdot\vec{S}_i \tag{1}$$

which can formally be mapped to a sG Hamiltonian.[15] The intrachain interaction $J/k=55$ K; $(J_{zz}-J)/J=-0.05$. The interchain coupling $J'\simeq10^{-3}$ J is not included. From these values one may estimate a crossover from Heisenberg to XY behavior[16] for

$$T<T_{CR}=0.44JS(S+1)/k=16\text{ K}\ .$$

The measurements reported here are performed at $T<7$ K and thus in the XY region. The ordering temperature of CHAB amounts to $T_c=1.50$ K (Ref. 13) at $B=0$.

The magnetic array consists of ferromagnetic $-Cu-Br_3-Cu-Br_3-$ chains running in the crystallographic c direction. The moments in each chain have a slight preference for the x direction in the easy XY plane where y coincides with the c axis. The x axis is located in the ab plane at an angle ϕ from the b axis. Two symmetry-related types of chains are present with $\phi=+25°$ and $\phi=-25°$, respectively. The anisotropy $(J_{xx}-J_{yy})/J$ within the easy XY plane has been determined by ferromagnetic resonance[14] (FMR) and amounts to 7×10^{-4}. Our experiments were performed in the paramagnetic state in magnetic fields along the a, b, and c axes. For both types of chains, the c axis coincides with the easy plane.

The specific heat of a single crystal of 2.5 g was measured by means of a heat-pulse calorimeter[13] with an accuracy of 2%. Temperature readings were obtained from a germanium thermometer and were corrected for magnetic field effects (at most 30 mK), if appropriate. Figure 1 shows the excess heat capacity ΔC obtained by subtracting the zero-field results from the data obtained from $B\parallel c$. In Fig. 2 the field dependence of ΔC is plotted. No demagnetization corrections were applied, since these amount to at most 50 G.

Although ΔC certainly includes the effect of B on the linear spin-wave excitations, this contribution on itself would yield a *decrease* of the specific heat with increasing field in the present region. Furthermore, a contribution arising from isolated magnetic ions (or impurities) can be ruled out.[8] Since, as will appear below, the NSLR measurements on this system also suggest a soliton contribution, we will now interpret ΔC in that light.

The soliton contribution to ΔC is[17]

$$\Delta C_s\simeq\frac{k}{a}\left(\frac{8}{\pi}\right)^{1/2}(16JS^2)^{-1}kT(E_s^0/kT)^{3/2}\times[(E_s^0/kt-\tfrac{1}{2})^2-\tfrac{1}{2}]\exp(-E_s^0/kT)\ , \tag{2}$$

where $E_s^0=8S(2Jg\mu_BBS)^{1/2}$, the soliton rest energy. With the appropriate values for CHAB ($E_s^0/k=10.85\sqrt{B}$), a good qualitative description of the data is obtained, but it seems that both the overall magnitude of ΔC and E_s^0 have to be adapted.

From Eq. (2) it appears that $\Delta C_S/kT$ is uniquely deter-

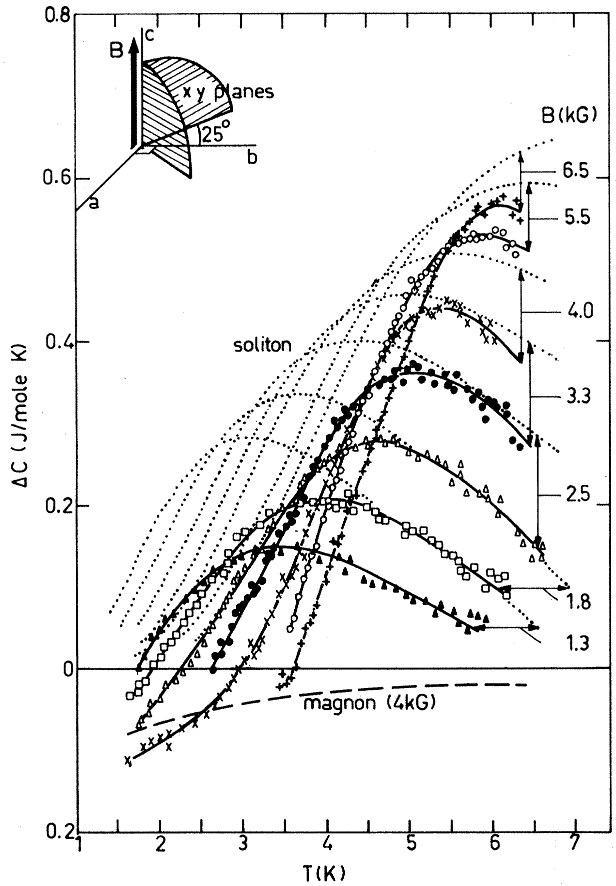

FIG. 1. Temperature dependence of ΔC. Drawn lines are guides to the eye. Dotted curves represent classical soliton theory with $E_s^0/k = 8.4\sqrt{B}$. The calculated magnon contribution is shown by a dashed curve. The inset shows the orientation of the easy XY planes.

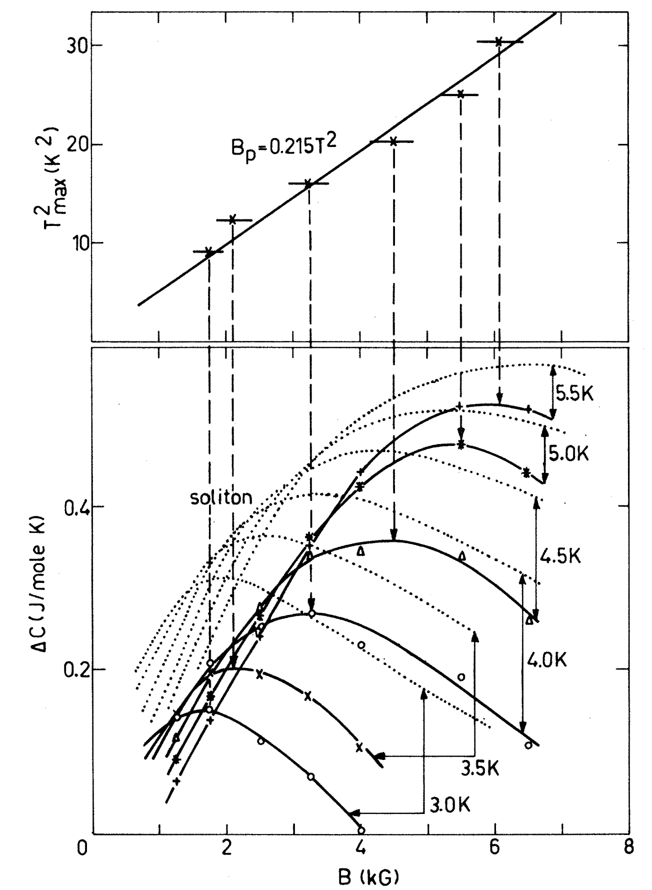

FIG. 2. Field dependence of ΔC, and the location of the maximum of $\Delta C(B)$. Dotted curves represent classical soliton theory with $E_s^0/k = 8.4\sqrt{B}$.

mined by E_s^0/kT and has a maximum for $E_s^0/kT \simeq 3.9$. If the data obey the relation $E_s^0 = \alpha\sqrt{B}$ predicted for a pure XY system, the maximum in $\Delta C(B)$ determines α through $B_{\text{peak}} = (3.9kT/\alpha)^2$. The data shown in Fig. 2 clearly reveal a quadratic dependence of B_{peak} on T and result in an experimental soliton energy $E_{s,\text{expt}}^0/k = 8.4\sqrt{B}$, indicating a reduction of $\sim 20\%$ compared with the classical value $E_s^0/k = 10.85\sqrt{B}$. This reduction may be compared with the prediction of Maki[11] which yields $E_s^0/k = 9.4\sqrt{B}$ for this case. The dotted curves in Figs. 1 and 2 are calculated with $E_{s,\text{expt}}^0$ and show a perfect qualitative agreement with respect to both the field and temperature dependence.

With respect to the overall magnitude of ΔC we recall that the data include the contribution of linear magnetic excitations. To illustrate the effect of this contribution we have included in Fig. 1 some results of a linear spin-wave calculation based on the field dependence of the energy gap observed by FMR.[14] The corrected magnitude of ΔC, however, is still too low. The reason for this discrepancy, which was also observed in $CsNiF_3$ (Ref. 8) and TMMC,[10] is yet unclear.

Measurements of ΔC were also performed with B along the b and a axes. The data for $B \parallel b$ almost coincide with those for $B \parallel c$. The data for $B \parallel a$ are shown in Fig. 3 for two representative values of B. Since a field $B \parallel a$ can be

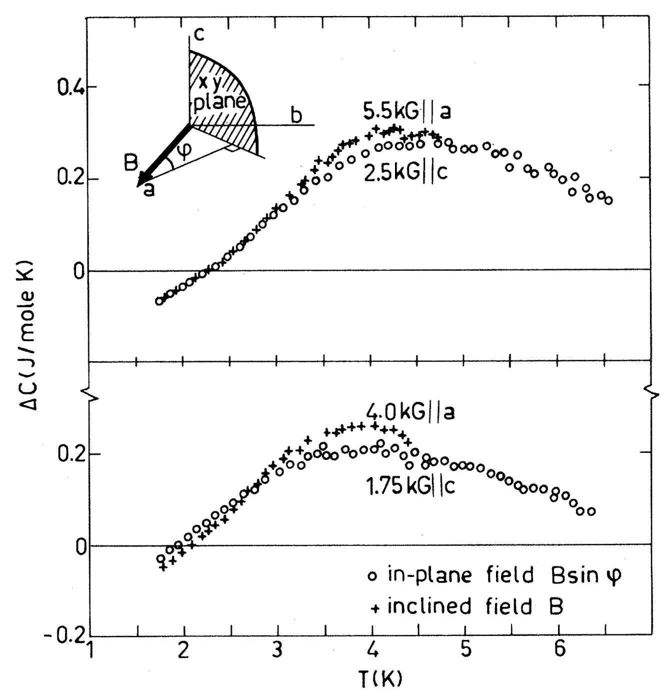

FIG. 3. Comparison of ΔC measured in inclined fields with ΔC measured in in-plane fields of magnitude B sinϕ.

decomposed into a component $B\sin\phi$ in the XY plane and a component $B\cos\phi$ along the real hard axis of the individual chains, we have also shown the data obtained in a field with a magnitude $B\sin\phi$ along c. This comparison reveals that the data for $B \parallel a$ can almost completely be explained by the effect of the in-plane component of the field. This observation corroborates an interpretation in terms of solitons since, as long as the out-of-plane component of the field is smaller than the out-of-plane anisotropy, only the in-plane component induces the symmetry breaking required for soliton excitations. Our results can be considered as the first experimental observation of this effect. One should note that the out-of-plane component of the field may alter the stability range of the solitons.[15,18] The present data, however, which have been collected at fields well below the in-plane critical field above which kink solitons are ustable (15 kG for CHAB),[15] do not indicate such an effect. Of course, the data should be corrected for the magnon contribution, but since this will not exceed $\Delta C \simeq -2\times 10^{-2}$ J/mole K in the present field and temperature range the conclusion of our analysis will remain unaltered.

The NSLR time T_1 has been measured in the paramagnetic phase, 1.2 K $< T <$ 10 K, in fields 1.5 kG $< B <$ 7 kG at nuclear hydrogen positions. The data were obtained on single crystals, using conventional pulsed NMR, with B applied along the crystallographic axes. The results for $B \parallel c$ are shown in Fig. 4 in reduced form. The drawn lines in the insert represent the result of a calculation of T_1 assuming linear spin-wave excitations. Although the qualitative behavior is similar to what is actually observed, especially at low T, serious deviations appear at higher temperatures. Therefore we will interpret the data on the basis of the soliton concept. When NSLR is due to collision with a one-dimensional soliton gas, the relaxation rate T_1^{-1} is[7]

$$T_1^{-1} \simeq \frac{1}{T}\exp(-E_s^0/kT) \simeq \frac{1}{T}\exp(-\alpha\sqrt{B}/T) \ , \quad (3)$$

where $E_s^0/k = \alpha\sqrt{B}$ is the soliton rest energy.

Equation (3) results in a linear relationship of $\ln(TT_1^{-1})$ and $\sqrt{B}/T$ for all fields and temperatures. As can be observed in Fig. 4, the data are located on a straight line for 1.8 K $< T <$ 8 K. At lower temperatures, where a spin-wave description is adequate, deviations occur. The data result in $E_s^0/k = 8.1\sqrt{B}$, which agree surprisingly well with the value $8.4\sqrt{B}$ obtained from the ΔC measurements.

Measurements of T_1^{-1} with B along the a and b axes were also performed. If only the in-plane component of B is considered, the data for $B \parallel b$ can also be described by Eq. (3). The data for $B \parallel a$, however, cannot simply be scaled by the same procedure.

In conclusion, we state that both the measurements of ΔC and T_1 reported here yield strong evidence of kink solitons in a ferromagnetic quantum ($S=\frac{1}{2}$) XY system. The field and temperature dependence of both T_1 and ΔC can very well be described by *classical* soliton theory, both for in-plane and inclined magnetic fields. This is surprising, since in the present quantum limit the validity of the mapping of the spin Hamiltonian to the sG Hamiltonian is not clear.[4] Although this question triggered part of the extensive theoretical effort,[19] which has been devoted to the properties of $S=\frac{1}{2}$ systems, general results for the infinite system in the presence of a symmetry-breaking field are not yet available.

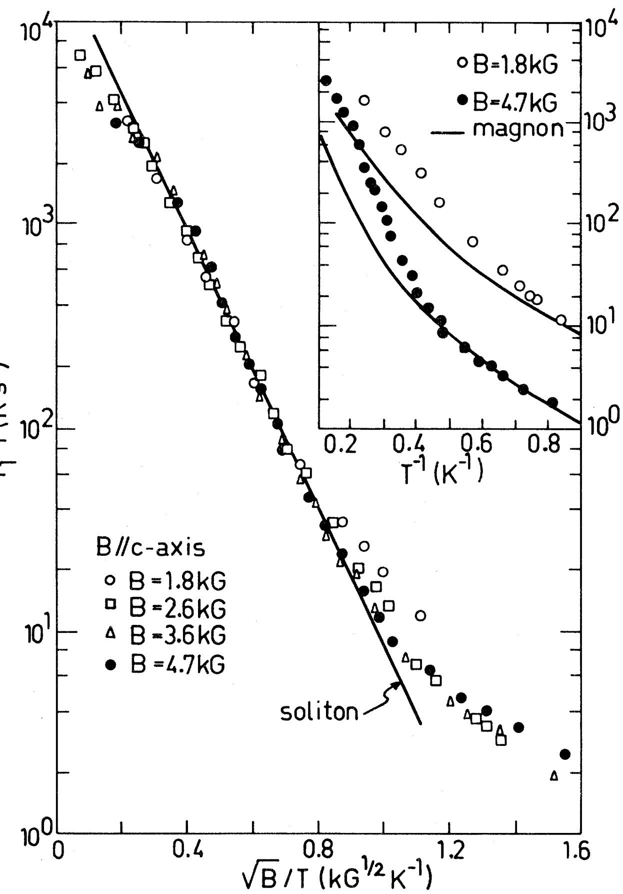

FIG. 4. Experimental behavior of $\ln(TT_1^{-1})$ as a function of $\sqrt{B}/T$. The solid line represents the classical soliton model with $E_s^0/k = 8.1\sqrt{B}$. The insert shows the prediction from linear spin-wave theory.

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[1] J. A. Krumhansl and J. R. Schrieffer, Phys. Rev. B 11, 3535 (1975).
[2] See, for instance, H. J. Mikeska, J. Appl. Phys. 52, 1950 (1980).
[3] J. K. Kjems and M. Steiner, Phys. Rev. Lett. 41, 1137 (1978).
[4] J. M. Loveluck, T. Schneider, E. Stoll, and H. Jauslin, Phys. Rev. Lett. 45, 1505 (1981).
[5] C. Reiter, Phys. Rev. Lett. 46, 202 (1981).
[6] M. Steiner, K. Kakurai, W. Knop, R. Pynn, and J. Kjems, Solid State Commun. 41, 329 (1982).
[7] T. Goto and Y. Yamaguchi, J. Phys. Soc. Jpn. 50, 2133 (1981).

[8]A. P. Ramirez and W. P. Wolf, Phys. Rev. Lett. 49, 227 (1982).
[9]J. Bouillot and W. G. Stirling, Solid State Commun. 33, 171 (1980); J. P. Boucher and J. P. Renard, Phys. Rev. Lett. 45, 486 (1980).
[10]F. Borsa, Phys. Rev. B 25, 3430 (1982).
[11]K. Maki, Phys. Rev. B 24, 3991 (1981); H. J. Mikeska, *ibid.* 26, 5213 (1982).
[12]C. P. Landee and R. D. Willett, Phys. Rev. Lett. 43, 463 (1979).
[13]K. Kopinga, A. M. C. Tinus, and W. J. M. de Jonge, Phys. Rev. B 25, 4685 (1982).
[14]A. C. Phaff, Ph.D. thesis, Eindhoven (unpublished).
[15]E. Magyari and H. Thomas, J. Phys. C 15, L333 (1982).
[16]A. C. Faria and A. S. T. Pires, J. Phys. C 12, 2637 (1979); F. Boersma, Ph.D. thesis, Eindhoven (unpublished).
[17]J. F. Currie, J. A. Krumhansl, A. R. Bishop, and S. E. Trullinger, Phys. Rev. B 22, 477 (1980).
[18]P. Kumar, Phys. Rev. B 25, 483 (1982); D. Hackenbracht and H. G. Schuster, *ibid.* 27, 572 (1983).
[19]J. D. Johnson and J. C. Bonner, Phys. Rev. Lett. 44, 616 (1980); T. Schneider and E. Stoll, *ibid.* 47, 377 (1981); M. W. Puga and H. Beck, J. Phys. C 15, 2441 (1982).