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OBSERVING SOLITONS IN ONE-DIMENSIONAL MAGNETIC SYSTEMS

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ABSTRACT

Classical models of one dimensional magnetic systems show that in addition to the linear spin wave excitations, there should exist localized, large amplitude excitations, that can move along the chains while retaining their integrity. It is expected that these excitations, "solitons", exist in real materials. We will discuss the progress that has been made to date in observing solitons in one dimensional magnets by means of neutron scattering, and the difficulties that still remain in unambiguously identifying the soliton contributions to $S(q, \omega)$.

INTRODUCTION

That solitons, mobile, localized non-linear excitations that retain their identity upon collisions, exist in classical models of one dimensional magnetic systems is clear mathematically^{1,2,3,4}. Considerable effort has gone into observing these excitations in real materials by means of neutron scattering, particularly TMMC and CsNiF_3 ^{5,6}, and it is my purpose to provide an introduction to the physics of solitons in magnetic systems and review the extent to which the efforts to observe them have been successful.

The theoretical interpretation of the results in these systems is based upon the sine Gordon equation. It is clear² that solitons exist in one dimensional magnetic systems that cannot be described by the sine Gordon equation, for instance, CsNiF_3 in the absence of a field.

THEORY AND EXPERIMENT FOR SINE GORDON-LIKE SYSTEMS

The sine Gordon equation

$$\frac{\partial^2 \phi}{\partial t^2} - \frac{1}{c^2} \frac{\partial^2 \phi}{\partial x^2} = \frac{m^2}{\beta} \sin \beta \phi \quad (1)$$

is an example of a completely integrable system. By means of non linear transformation, an arbitrary solution may be regarded as being composed of three distinct types of excitations. These are unbounded (in space) oscillatory solutions closely related to the phonons of the linearized theory, and going over to these phonons as their amplitude decreases, bounded oscillatory solutions of finite amplitude that have envelopes that can move along the chain, called breathers, and fixed amplitude kink solutions, corresponding to a 2π change in ϕ , that are also free to move along the chain.

These latter are usually called solitons, and will be so denoted in the rest of the paper. The large amplitude breathers may be thought of as bound states of a kink and anti-kink, while the small amplitude ones can be thought of as bound states of a pair of phonons. The solitons of the theory are sometimes called topological solitons. The limit of ϕ as $x \rightarrow \pm\infty$ is different for such solitons. They are distinguished from pulse solitons, such as the breathers, for which the system returns to the same state as one passed through the soliton. It is characteristic of the system that what one regards as the elementary excitation and what composite depends upon the perspective one wishes to adopt. This is even more apparent in the quantum version of the model.

Classically, the parameter β is irrelevant. By defining $\phi' = \phi\beta$, it could be eliminated from the equation of motion. The Hamiltonian for the system is

$$\mathcal{H} = \frac{\hbar}{c} \int dx \left[\frac{1}{2} c^2 \phi_x'^2 + \frac{1}{2} \phi_t'^2 - \frac{m^2 c^2}{\beta^2} \cos \beta \phi \right] \quad (2)$$

and in calculating the thermodynamics, this scaling would simply change the effective temperature. In the quantum theory, however, $\hbar$ sets an intrinsic scale for the variations of ϕ , and the magnitude of β is an essential parameter of the theory, determining the nature of the spectrum by providing a measure of the anharmonicity^{8,9}. For $\beta^2 \ll 4\pi$, the spectrum consists of a ground state, whose energy is mc and that corresponds to a phonon, a series of excited states corresponding to the breathers, and finally a state corresponding to an unbound kink anti-kink solution at an energy of $16 mc/\beta^2$. As β increases, the number of breather states decreases, until at $\beta^2 = 4\pi$, there are none. For TMMC and CsNiF_3 , it turns out that $\beta^2 \approx 7$. The smaller the value of β , the greater the correspondence between the classical solutions and the quantum theory, and it is appropriate to use the classical theory as a first approximation in interpreting the experiments on TMMC and CsNiF_3 .

The calculation of the contribution to correlation functions from the solitons in the classical model was done first by Kawasaki¹⁰ in the limit of a dilute gas of solitons, and this has been extended by Mikeska¹¹ and Allroth to include the effect of the presence of the phonons on the soliton contribution at higher temperatures. Both of these calculations contain heuristic elements, and it is not clear whether they are in fact exact, even asymptotically as the temperature approaches zero. We shall be interested in the extent to which they are confirmed by experiment.

Neither CsNiF_3 nor TMMC have Hamiltonians that are obviously of the sine Gordon form, which is to be regarded as an effective Hamiltonian, valid at low temperatures. The correspondence, due first to Mikeska^{3,4} is most easily seen for CsNiF_3 , which is

describable by the Hamiltonian

$$\mathcal{H} = -J \sum_i \vec{S}_i \cdot \vec{S}_{i+1} + DS_i^Z{}^2 - g\mu_B H \sum_i S_i^x \quad (3)$$

with $S=1$.

At low temperatures, i.e. $KT \ll \sqrt{DJ} S^2$, the spins will lie preferentially in the x,y plane. If we identify ϕ as the angle of departure of the spin direction (for the classical system) from the x axis in the x-y plane, and θ as the angle out of the plane, and observe further that ϕ and S_Z are canonical variable, so that $\dot{\phi}_1 = (\delta H)/(\delta S_Z) = (2S_i^Z)/(D)$, we find in the continuum limit, and to lowest order in θ ,

$$\mathcal{H} = \int \frac{dx}{a} \left[JS^2 a^2 \frac{1}{2} (\phi_x)^2 + \frac{1}{4D} \dot{\phi}^2 - g\mu_B H S \cos \phi \right] \quad (4)$$

which is the sine Gordon Hamiltonian with

$$c^2 = \frac{2DJS^2 a^2}{h^2}, \quad m^2 = \frac{g\mu_B H}{JSa^2}, \quad \beta^2 = \sqrt{\frac{2D}{JS^2}}$$

The antiferromagnet requires a slightly more complicated derivation and differs in a fundamental way. The spins are nearly perpendicular to the field, most of the time, and a soliton corresponds to a rotation by π , not 2π . As a consequence, the soliton connects regions of the lattice that are in the two inequivalent ground state configurations. For the ferromagnet, the two configurations on either side of the soliton are identical. For a fixed density of solitons in a chain, the disordered region in the ferromagnet extends only over the distance occupied by the solitons, and vanishes with the density. For an antiferromagnet, the entire chain is always disordered, and it is the inverse coherence length that vanishes with the density. Thus while the solitons show up in the ferromagnet as a decrease in the intensity of the Bragg plane and a contribution to the inelastic scattering, in the antiferromagnet, they can be seen in the modification of the Bragg plane into a lorentzian whose width in q is basically the inverse separation of the solitons and whose width in frequency is determined by the mean time it takes the solitons to move that spacing. It is much easier, as a result, to observe the effect of the solitons in the antiferromagnet, although in a sense, what one is observing is primarily the space between the solitons.

In the sine Gordon approximation, $S^x = S \cos \phi$, $S^y = S \sin \phi$ and the

prediction of the theory of Kawasaki and Mikeska for

$$S_q^{\alpha\alpha}(\omega) \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle S_q^{\alpha}(t) S_{-q}^{\alpha} \rangle e^{i\omega t} dt \quad \text{is}$$

$$\left\{ \begin{matrix} \text{xx} \\ \text{yy} \end{matrix} \right\} S_q(\omega) \equiv \frac{64}{\pi} \frac{1}{KT} \frac{1}{cq} e^{-\frac{8mJa}{KT}} \left(1 + \frac{1}{2} \frac{\omega^2}{(cq)^2} \right) \left\{ \left(\frac{\pi q}{2m} / \sinh \frac{\pi q}{2m} \right)^2 \right. \\ \left. \left(\frac{\pi q}{2m} / \cosh \frac{\pi q}{2m} \right)^2 \right\} \quad (5)$$

The factors in curly brackets are the form factors for the projections of the spin component of a single soliton on the x and y axes.

The integrated intensity is proportional to these factors and so provides a direct measure, within this theory, of the shape of the soliton. The density of solitons in this theory is proportional to $e^{-E_s/KT}$, where $E_s = g\mu_B HS$. Boucher et al. have shown quite convincingly that the width in $q(\Gamma_s)$ of the Bragg plane, and its width in $\omega(\Gamma_D)$ are proportional to such an exponential in the region of low soliton density, where the theory is expected to apply. The value for E_s that they obtain differs significantly from its classical value, which may be due to quantum effects¹². They have also obtained another measure of Γ_D from NMR measurements of T_1^{-1} , and it agrees with the neutron scattering results.

Of course there is no genuine Bragg plane in a one dimensional system, and in the absence of a field, the widths Γ_D and Γ_s (the lineshape is not actually lorentzian) can be calculated from a theory that includes only the linear excitations, the spin waves¹³. It remains an unsolved problem to interpolate between the case that $H=0$ and large enough H that the soliton theory can be expected to apply, both conceptually and with a practical calculation.

The problem of distinguishing between the contribution to the correlation function that arise from the linear excitations, the spin waves, and the non-linear excitations, the breathers and solitons, is more serious in the ferromagnet. In fact, it seems unlikely that a clear theoretical distinction between the spin wave contribution and the breather contribution can be made at all, as both contributions can presumably be obtained in perturbation theory in the parameter β . The soliton contribution, proportional

to $e^{-\frac{8mg}{\beta KT}}$ is non-analytic in β , and is theoretically distinguishable, but the problem is to separate the soliton contribution experimentally from the anharmonic spin waves. The original experiments interpreted the inelastic scattering seen near $\omega=0$ at small wavevectors as being due to scattering off a gas of solitons, the spread in frequencies being essentially the doppler

shifts from the moving solitons. The variation of the intensity of this central peak with q is predicted by the theory of Kawaski and Mikeska to correspond to the structure factor for a single soliton, so that these observations appeared to provide very direct observation of the solitons in the chain. It soon became clear that the interpretation was not so simple.

Scattering from pairs of spin waves, or in other words, fluctuations of the length of the spin component along the field, provides an additional mechanism for producing a central peak. (In fact, it produces also scattering at twice the spin wave frequency, which has now been observed¹⁴.) The variation of the intensity with field temperature and wavevector, are all well described by the two spin wave scattering¹⁵. It has also been argued from simulations, that the sine Gordon approximation is not valid at the temperatures and fields that the experiments were done¹⁶. Allroth and Mikeska have argued that the data is best fit by a sum of the two contributions, the two being roughly equal at the highest temperatures of observation, but the uncertainty in the absolute value of the observed intensity, which could presumably be eliminated by comparing with the spin wave intensities prevents any unambiguous statement from being made. They do indicate that at least for the spin wave intensities and central peak structure factor, that the sine Gordon theory is a satisfactory approximation at the temperatures and fields of interest.

To resolve these difficulties in interpretation, Steiner¹⁵ et al. have done an experiment in which the scattering vector is aligned along the field, so that the two spin wave scattering in $S_{q,q}^{xx}(\omega)$ does not contribute. They observe additional intensity q around $\omega=0$ in this case as well, which they attribute to the presence of solitons. There is, however, an anharmonic contribution to this central peak intensity that comes from the spin waves.

The spin operator has a boson equivalent that, in leading order that contains the anharmonic effect, is

$$S_i^y = i\sqrt{S}(a - a^\dagger) - i \frac{1}{2\sqrt{2S}}(a_i^\dagger a_i a_i - a_i^\dagger a_i^\dagger a_i) \quad (6)$$

The second term contributes to the scattering a term which is, neglecting the anharmonicity in the Hamiltonian,

$$\frac{1}{8S^N} \sum_{q_1 q_2 q_3} \Delta(q - q_1 - q_2 - q_3) \left| r_{q_1 q_2 q_3} \right|^2 \{ n_{q_1} (n_{q_2} + 1)(n_{q_3} + 1) \delta(\omega - \omega_{q_1} + \omega_{q_2} + \omega_{q_3}) + n_{q_2} n_{q_3} (n_{q_1} + 1) \delta(\omega + \omega_{q_1} - \omega_{q_2} - \omega_{q_3}) \} \quad (7)$$

where $r_{q_1 q_2 q_3} = \alpha_{q_1} \alpha_{q_2} \alpha_{q_3} + \beta_{q_1} \beta_{q_2} \beta_{q_3} + 2\alpha_{q_1} \beta_{q_2} \beta_{q_3} + 2\beta_{q_1} \alpha_{q_2} \alpha_{q_3}$

The α_q , β_q are the coefficients of the Bogolubov transformation, the n_q occupation numbers and the ω_q the spin wave frequencies, all defined in ref. 15.

This is not the only anharmonic contribution, as there are terms coming as well from the anharmonic terms in the Hamiltonian that could also be considered. We expect them to make comparable contributions, and a rigorous calculation has not yet been done. For the parameters of the experiment, the solitons do give a significant contribution to the scattering near $\omega=0$, and the three spin wave scattering is considerably smaller. The integrated intensity of the solitons is proportional to the structure factor and has a peak at $q \approx .06$ R.L.O. Much of the three spin wave scattering would be included in the spin wave cross section, with the procedure used by Steiner et al. for fitting the data, and the intensity of the remainder would appear as a nearly q independent background. We conclude that the q dependence of the intensity observed cannot be explained by the lowest order spin wave theory. The possibility remains that a more accurate calculation could explain it, and we think this unlikely. We would conclude that the soliton interpretation of scattering near $\omega=0$ is tenable.

Nevertheless, the situation in CsNiF_3 is far from satisfactory. The observed intensity in the central peak, in $S_{YY}(\omega)$ is about a factor of three¹⁸ above what one expects from the lowest order soliton theory and the finite temperature corrections serve to reduce the theoretically predicted intensity significantly. If we take the experiment at face value, then we must conclude that the prefactors in the theoretical expression (5) are significantly in error.

A further test of the soliton theory in CsNiF_3 has been suggested. It has been observed by both Kumar¹⁹ and Magyari and Thomas²⁰ that the solitons become unstable if the field exceeds 18 Kg. This is because the spins that are antialigned with the field can lower their energy by tipping out of the plane if the field is large enough, and 18 Kg is the boundary for this to occur. Magyari and Thomas argue that this should lead to characteristic singularities in the soliton contribution to the correlation function, and it would be very interesting to determine if these were observable.

We have been discussing so far x-y like systems, in which an external field breaks the symmetry. In Ising like systems, for which the sign of D in (3) is negative, Mikeska has shown that the classical continuum limit for the antiferromagnet leads again to the sine Gordon equation⁴ the angle now being the deviation of the spin direction from the Ising axis. The solitons connect two distinct ground states, and so disorder the system in a manner similar to the x-y antiferromagnet. The external field can be zero. If it is not, a double sine Gordon equation results, with terms involving both $\cos\phi$ and $\cos 2\phi$.

A quantum system, $C_5C_0CL_3$ exists, with spin $1/2$, that has the symmetry, and furthermore, the exchange terms are sufficiently small as to be treatable by perturbation theory. The basic excitation is indeed a moving domain wall, as first pointed out by Villain²¹. Extensive experimental work, in the absence of a field has been done by Yoshikawa, Hirakawa, Satija and Shirane²² and both numerical calculations using finite chains and analytic calculations using perturbation theory have been done by Ishimura and Shiba²³. Together with the earlier analytic calculations of Villain that are valid for wavelengths short compared to a coherence length, these provide a clear picture and a quantitative understanding of many aspects of the system.

The transverse excitation spectrum can be interpreted as being due to exciting pairs of domain walls. The degree of freedom corresponding to the relative motion of the pair leads to a band of excitations rather than a definite relationship between ω and q . The spectral density in the band is more asymmetric than predicted, perhaps due to interchain coupling. The longitudinal scattering is primarily a band around $\omega=0$ that can be thought of again as the doppler shifting of the neutrons due to scattering off of individual domain walls. The theory is in satisfactory agreement with experiment if $q > K$, but the long wavelength phonons are sensitive to multiple domain walls, and there is no analytic theory available.

We want to emphasize that the theory for the system is very much a quantum theory,, and although it seems natural to associate the excitations observed in $C_5C_0CL_3$ with the classical solitons, this is a purely heuristic identification at this point. $\beta^2=8/S$ for the mapping onto the sine Gordon theory, and for spin $1/2$, the quantum sine Gordon theory is still definable,, although there are no spin waves or breathers, since $\beta^2 > 4\pi$. This is in accord with the theory based directly on the spin Hamiltonian. Presumably, a system with $S=1$ would show both spin waves and solitons.

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