Elementary Excitations in Quantum Antiferromagnetic Chains: Dyons, Spinons and Breathers.

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Considering experimental results obtained on three prototype compounds, TMMC, CsCoCl\textsubscript{3} (or CsCoBr\textsubscript{3}) and Cu Benzoate, we discuss the importance of non-linear excitations in the physics of quantum (and classical) antiferromagnetic spin chains.

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I. INTRODUCTION:

In the last twenty years, various experimental and theoretical studies have been devoted to antiferromagnetic (AF) chains. One fascinating problem concerns the nature of the elementary excitations in such systems. As a crucial ingredient, one has to refer to the concept of non-linear excitations (NLE). Typical examples of NLE are provided by the solitons and the breathers. These particular excitations are solutions of the sine-Gordon equation, which, in the wide field of the NLE, is to be considered as a prototype model \[\text{[1]}\]. The first evidence of fluctuations associated with solitons in AF chains was obtained on a classical spin system (in the 80’s) \[\text{[2]}\]. Recently, evidence for breathers was experimentally obtained on a quantum spin system \[\text{[3]}\]. A short review is presented, along which, the role of the NLE in AF chains is discussed. Classical as well as quantum spin systems are analyzed. The case of anisotropic spin models is considered and a relation with the isotropic case is proposed. Quantization effects are also shown to complete the descriptions of the NLE in AF chains: for the solitons, this leads to another NLE concept, the dyons; for the breathers to a discretization of the excitation spectrum. Along this paper, we shall refer explicitly to three compounds: (CH\textsubscript{3})\textsubscript{4}NMnCl\textsubscript{3}, alias TMMC, two Co compounds, CsCoCl\textsubscript{3} and CsCoBr\textsubscript{3} \[\text{[4]}\], and Cu(C\textsubscript{6}H\textsubscript{5}COO)\textsubscript{2}.3H\textsubscript{2}O, alias Cu Benzoate \[\text{[5]}\]. All of them are known for very long to be good models of 1-dimensional antiferromagnetic chains. In TMMC, the spin value is large, \(s = 5/2\), and a semi-classical spin description is a good approach. The other compounds, however, are representative of quantum spin systems, with \(s = 1/2\).

II. TMMC:

TMMC is known to provide examples of quasi-isotropic Heisenberg AF chains. At \(T_{\text{co}} \simeq 20\) K, however, because of the small dipolar anisotropy \(D\), a crossover occurs, which changes the spin system from isotropic to easy-plane behaviors \[\text{[6]}\]. The spins can be viewed as being repelled in the XY plane perpendicular to the chain Z direction. Then, if one applies a magnetic field \(H\) in the XY plane - along the Y axis, for instance - another crossover occurs because of the preferred spin-flop configuration. In this later phase, the spins are mainly aligned along the X axis: they form an Ising-like AF state where the soliton regime takes place. The soliton phase diagram for TMMC is shown in Fig. 1 \[\text{[4]}\].

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Soliton phase diagram of TMMC \[\text{[4]}\]. \(H_c\) is the cross-over field between the soliton regimes A and B (see the insets).}
\end{figure}
Two soliton regimes (A and B in Fig. 1, see also the insets) can be defined. Both they correspond to the sine-Gordon model. The difference between A and B results from the relative “competition” between the two effective anisotropies, one due to $D$, the other induced by $H$.

We mainly limit the discussion to the “transverse” soliton regime A. For TMMC, the initial Hamiltonian is given by:

$$\hat{H} = \sum_n 2J s_n s_{n+1} - 2D s_n^z s_{n+1} - g\mu_B H s_n^z$$  \hspace{1cm} (1)$$

with $J \simeq 6.8$ K and $D \simeq 0.16$ K and $s = 5/2$. Each spin can be described as a vector, the orientation of which is defined by two angles, $\theta$ and $\phi$. In configuration A, the spins are assumed to remain within the XY plane. Accordingly, $\theta$ remains fixed to the value $\pi/2$, and $\phi$ is defined by the angle between the spin and the external field $H$. In the Ising-like phase of TMMC, the spin vectors is simply written as $s_n^z = (-1)^n \cos(\phi)$. Within a classical description (and in the continuum limit), and after the variable change $\psi = \pi - 2\phi$ is made, hamiltonian (1) is changed into

$$\hat{H} = J s^2 / 2 \int_{-\infty}^{+\infty} dz / 2 \left[ \frac{1}{C^2} \left( \frac{\partial^2 \psi}{\partial t} \right)^2 + \left( \frac{\partial \psi}{\partial z} \right)^2 + m^2 \left( 1 - \cos(\psi) \right) \right]$$  \hspace{1cm} (2)$$

which is that of the sine-Gordon model. Here, $C = 4 Js[1 - D/(2J)]^{1/2}$ is the soliton velocity and $m = g\mu_B H/4Js$ defines the soliton mass. The elementary excitations of this model are typical non-linear excitations (NLE): the solitons (and antisolitons) and the breathers. A soliton in TMMC (or an antisoliton) can be viewed as a domain wall (extending over several lattice spacings), with a well-defined shape as represented in Fig. 2. The breather modes are soliton-antisoliton bound-states. As a general property, when NLE move along the chains, they maintain an exact balance between two energy contributions, the “potential” and the “internal” energies defined by the two last terms of (2), respectively. This explains the remarkable “integrity” of the NLE excitations.

![Fig. 2. Schematic representation of a (broad) soliton in TMMC.](image)

As can be seen in Fig. 2, inside a soliton, the spins undergo a $\pi$ rotation (this provides an alternative and a more general definition of solitons in AF chains). By looking at the fluctuations induced by the soliton motion, an accurate description of the soliton regimes in TMMC has been obtained. Concerning the breather excitations, the situation is experimentally more complicated. In spin systems, a discretization of the breather spectrum is to be expected, which, then, should result in a set of discrete excitation branches, hereafter denoted $B_1$ ... $B_n$. In this quantization process, the lowest breather $B_1$ coincides with the usual magnon excitation.

If we consider only the A soliton regime, the lowest breather gap is that of the lowest magnon branch. The experimental gap value is in agreement with the value predicted by (2): $E_1^A = g\mu_B H$. In TMMC, however, there exist two competitive anisotropies as mentioned above. Accordingly, there is another magnon branch with another gap: $E_1^B = 4S\sqrt{D/J}$ ($\simeq 0.8$ meV). This higher gap is that of the lowest breather branch associated with the B soliton regime (see the insets in Fig. 1). In the quantization process, the breathers must be considered as a whole. As a result, additional peaks can be expected in the breather spectrum. This agrees with the observation of “unexpected” peaks at energies $E \simeq E_1^B \pm E_1^A$.

III. THE CO COMPOUNDS: CSCOCL$_3$ AND CSCOBR$_3$:

An alternative soliton (or kink) model for AF chains has been proposed by J. Villain, in 1975. It applies to the following quantum ($s = 1/2$) Ising-like Hamiltonian:

$$\hat{H} = 2J \sum_n s_n^x s_{n+1}^x + \varepsilon (s_n^x s_{n+1}^y + s_n^y s_{n+1}^x)$$  \hspace{1cm} (3)$$

with $\varepsilon < < 1$. For this simple model, the groundstate is the Néel state as represented in Fig. 3a.

The first excited state is displayed in Fig. 3b: it contains a narrow domain wall (extending over only one lattice spacing) and, as expected, it is associated with a $\pi$
rotation of the sublattices. For this model, the dispersion of the soliton branch is shown in Fig. 4a.

![Diagram](image1)

FIG. 4. a) Dispersions of the soliton excitations in the Villain model: the solid line is for a small \( \varepsilon \) value; the dashed line, for a large \( \varepsilon \) (\( \to 1 \)). The arrows illustrate experimental transitions (see text). b) The fluctuation spectrum of the (individual) soliton mode (in zero field).

In the same figure, the arrows show the transitions to be realized in experiments. The transitions induced from the ground state (the dash arrow) are forbidden. They would require that an infinite number of spins be simultaneously flipped (in order to realize the \( \pi \) rotation of the sublattices). This establishes a pertinent result for Ising-like AF chains: the dispersion of a soliton branch cannot be determined experimentally. Only transitions inside the soliton branch (shown by the full arrow) are possible: they describe the fluctuations of (individual) solitons when they move along the chains. The corresponding spectrum is drawn in Fig. 4b: the fluctuations are seen to develop at very low energy (around zero energy). A peak (i.e., a single “maximum”) in the fluctuation spectrum is expected at \( E_{\text{max}}(q) = 4\varepsilon J \sin(q) \).

![Diagram](image2)

FIG. 5. Zeeman splitting of the soliton dispersion in the Villain model for a field parallel (a) and transverse (b) to the Ising direction.

Such a kink in quantum spin chains is, in fact, a degenerate state. As shown in Fig. 3c, it can be defined in two ways, which both agree with the \( \pi \) rotation defined above. They differ, however, by the spin value (\( \pm 1/2 \)) inside the domain wall. A kink, i.e., a soliton, is a doublet state, entirely defined by the additional quantum spin number \( S = 1/2 \). Accordingly, in a field \( H \), a Zeeman splitting occurs in the dispersion, as shown in Figs. 5a and b. This splitting is different for \( H \) applied parallel (\( H_\parallel \)) and perpendicular (\( H_\perp \)) to the Ising axis \( Z \). The allowed transitions are now induced between the two split branches (the full arrows in Figs. 5). Finally, this splitting results in a “doubling” of the observable soliton modes with two maxima, as shown in Figs. 6a and b.

![Diagram](image3)

FIG. 6. Doubling of the soliton modes in the Villain model, for \( H \) parallel (a) and perpendicular (b) to the Ising direction.

The Co compounds, CsCoCl\(_3\) and CsCoBr\(_3\), are good examples of the Villain model, with \( J \approx 75 \) K and \( \varepsilon \approx 0.12 \). The fluctuations associated with the solitons have been clearly identified. In particular, with CsCoBr\(_3\), the low-energy fluctuations characterizing the (individual) soliton modes have been observed by neutron inelastic scattering (NIS)\(^\text{[13]}\). Evidence for a “maximum” in the fluctuation spectrum requires a high instrumental resolution. Measurements have been performed at the Institut Laue-Langevin (ILL, Grenoble, France) using the high-flux high-resolution spectrometer IN14\(^\text{[14]}\). An example of the soliton mode observed in zero field for the wavevector transfer \( \Delta k = q = \pi/2 \) is displayed in Fig. 7a. A single “maximum” is clearly detected. Its position agrees well with the expectation (the full line is a theoretical curve which takes into account the instrumental resolution, \( \delta E \approx 1 \) meV). In Fig. 7b, the same energy scan performed in presence of a magnetic field (\( H \approx 9.82 \) T) is reported. Two maxima are now visible: this result (the full line is the theory) establishes the predicted doubling of the soliton modes.

The neutron data reported in Fig. 7 correspond to magnetic transitions associated with the particular momentum transfer \( \Delta k = q = \pi/2 \) (the full arrows in Figs. 4a and 5). Transitions with \( \Delta k = q = 0 \) (the vertical dash arrows in Fig. 5) can also be considered. They are realized in an ESR experiment. Such measurements have been performed on the compound CsCoCl\(_3\)\(^\text{[15]}\). They are referred to as Soliton Magnetic Resonance (SMR). Examples of SMR signals are reported in Fig. 8, for
both $H_\parallel$ and $H_\perp$. As shown by the full lines (theoretical curves), the SMR data agree also very well with this concept of a $S = 1/2$ soliton state.

IV. QUANTUM ISOTROPIC SPIN CHAINS:

Up to now, we have considered Ising-like chains, i.e., anisotropic systems, where the NLE are rigorously defined. Within the same context, we may try to reach a physical picture for isotropic chains. We follow the discussion presented by Haldane for classical spin systems [16], but we apply it to the quantum case of the Villain model. Let us consider the change occurring in the soliton dispersion while the anisotropy is slowly removed, i.e., when $\varepsilon \to 1$ in Eq. 3. As shown by the dash line in Fig. 4a, the amplitude of the dispersion increases, and at $\varepsilon = 1$, i.e., the isotropic point, the gap of the soliton branch would close (at the momentum value $k = \pi/2$). A phase transition occurs [13], which can also be viewed as an isolant-metal transition [19]. The dyon states (i.e., the pairs of $+1/2$ and $-1/2$ solitons) are seen to fill in the groundstate of the new phase, which is that of the quantum isotropic Heisenberg Hamiltonian

$$ \hat{H} = \sum_n 2JS_n s_{n+1} $$  \hspace{1cm} (4)

For this model, another concept, the spinon concept, is commonly used [20]. A spinon is defined as an “entity” associated with a $\pm 1/2$ spin value and the groundstate of this isotropic Hamiltonian is made of pairs of spinons. Such pairs of spinons compare well with the $\pm 1/2$ dyon states defined above.

The description of the properties of quantum Heisenberg chains started with the Bethe ansatz, in 1931 [21]. In the 60’s, based on the Jordan-Wigner transformation, many studies were referring to a model of strongly-interacting fermions [22]. In the 70’s, more sophisticated analyzes were proposed [23], which yields the modern field-theoretic derivations, which in the recent years, have renewed and enlarged our understandings of so many quantum spin systems. Cu Benzoate is such a recent example. For this compound, one is led to refer also to the sine-Gordon model [24].

In anisotropic systems, the spin direction is easily defined (for instance, by the angle $\phi$ in Fig. 2), and/or with respect to the groundstate. The pertinent variable in the sine-Gordon Hamiltonian of Eq. 2 remains this angle $\phi$. In isotropic systems, however, there is no definite direction for the spins. Alternative descriptions have been proposed [23]. Usually, they starts with the Jordan-Wigner transformation. This leads to a fermionic representation of the Hamiltonian with, in reciprocal space, the fermions operators $\alpha_q$ ($\alpha_q^+$) and $\beta_q$ ($\beta_q^+$). It is then recognized that the significant physical quantities are not contained in the fermion operators but rather in the fermion densities: $\rho_\alpha \approx \sum_k \alpha_{k+q}^* \alpha_k$ and $\rho_\beta \approx \sum_k \beta_{k+q}^* \beta_k$ (this is the so-called “bozonization”). Another transformation - this is the basis of the field theoretic approach - brings back the equations to the real space where a field “$\psi$” is
defined. Applying this procedure to the isotropic hamiltonian (4) leads to

$$\hat{H} = J/8 \int_{-\infty}^{+\infty} dz/2 \left[ 1/\bar{C}^2 (\partial \bar{\psi}/\partial t)^2 + (\partial \bar{\psi}/\partial z)^2 \right]$$

(5)

with $\bar{C} = \pi J$. In this expression, the field $\bar{\psi}$ does not compare to the angle $\psi$ introduced in Eq. 2: $\bar{\psi}$ is related to the fermion densities $\rho^+$ and $\rho^-$ defined above. Eq. 5 is not the sine-Gordon Hamiltonian. The last term which, in Eq. 2, introduces the non-linearity is missing. Accordingly, the excitations corresponding to Hamiltonian (5) follow linear dispersions (i.e., no energy gap). They describe the lowest part of the spinon continuum, which characterizes the excitation spectrum of (4).

As it is known, the application of a field $H$ on hamiltonian (4) leads to

$$\hat{H}_{\text{st}} = \hat{H} + \sum_n 2J s_n s_{n+1} - g_\mu B H s_n^2 - g_\mu_B h_{\text{st}} s_n^2$$

(6)

with $J \simeq 5.8$ K, and where $h_{\text{st}}$ is proportional to the applied field: $h_{\text{st}} \simeq 0.09H$ (for $H$ applied parallel to the c axis) [28]. The field-theoretic derivation of this hamiltonian [24] transforms Eq. 6 into the sine-Gordon equation:

$$\hat{H} = J/8 \int_{-\infty}^{+\infty} dz/2 \left[ 1/\bar{C}^2 (\partial \bar{\psi}/\partial t)^2 + (\partial \bar{\psi}/\partial z)^2 + \bar{m}^2(1 - \cos(\bar{\psi})) \right]$$

(7)

where $\bar{C}$ and the field $\bar{\psi}$ are defined in a similar way as in Eq. 5. The soliton $\bar{\psi}$ is now a function of the staggered field: $\bar{m} \simeq 1.85(h_{\text{st}}/J)^{2/3} / \ln(h_{\text{st}}/J)$ [25]. The low-energy excitations of (7) are solitons (antisolitons) and breathers.

V. CU BENZOATE:

In the 70’s, CuBenz was considered as a good example of quantum isotropic AF chains [25]. Recently, however, it has been observed that, in a field, an energy gap opens in the energy spectrum [26], in contradiction with the isotropic hamiltonian (4). As it is known, the application of a field $H$ on hamiltonian (4) develops a “dynamical incommensurability”: the wavevectors where the critical zero-energy fluctuations take place are shifted by $\delta q_{\text{inc}} = 2\pi \sigma$, where $\sigma$, which is the magnetization per spin, is an increasing function of $H$. The dispersions, however, remain linear at low energy and no energy gap is induced by a field.

FIG. 9. A model for the dispersion of the elementary excitations in Cu Benzoate: the soliton/antisoliton branch gives a gap at $q = 0$ and at the incommensurate wavevector $q = \pi - \delta q_{\text{inc}}$. The breather branches give a gap at $q = \pi$ and at the incommensurate wavevector $q = \delta q_{\text{inc}}$. The dashed arrows corresponds to NIS [26], the full arrows to ESR [3].

In Fig. 9, a complete representation of the elementary excitations in Cu Benzoate is proposed. The bold lines correspond to the sine-Gordon predictions (low-energy limit). In this figure, three breather branches are represented: $B_1$, $B_2$ and $B_3$.

We now review briefly the recent experiments performed in Cu Benzoate. In Fig. 9, the transitions observed by NIS are shown by the dash arrows (these measurements have been performed at a single field value $H \simeq 7$ T) [26]. The dynamical incommensurability has been observed at $q = \pi - \delta q_{\text{inc}}$. At this precise wavevector value, the gap of the soliton/antisoliton branch is directly measured. At $q = \pi$, the two first breather modes, $B_1$ and $B_2$, have been observed [29]. Recently, ESR measurements have also been performed in Cu Benzoate [30]. In Fig. 9, the transitions observed by ESR are shown by the vertical dotted and solid arrows. The solid arrow shows that the soliton/antisoliton gap is measured at $q = 0$. The dotted arrow probes the breather modes, also at $q = 0$ [30]. All these ESR measurements have been performed in a wide field range, up to $H \simeq 20$ T. The observed field dependence of the soliton gap is displayed in Fig. 10. In low field, the agreement with the sine-Gordon model (the full line) is very good: $E_G = \bar{m} J \simeq H^{2/3}$. An appreciable deviation, however, occurs in high fields. In Fig. 11, the field dependence of the three first breathers,
$B_1$, $B_2$ and $B_3$ as probed by ESR (i.e., at $q = 0$) is obtained in the full field range.

FIG. 10. Field dependence of the soliton gap observed by ESR (Voigt configuration) in Cu Benzoate.

VI. CONCLUSION:

With these compounds - i.e., TMMC, the Co compounds and Cu Benzoate - the importance of the NLE in the low-T properties of AF chains is well exemplified. They are seen to play a crucial role in both anisotropic and isotropic systems. They come in the description of both quantum and classical spin chains. TMMC and Cu Benzoate are both well described by the sine-Gordon model. Fundamental differences, however, occur between the two systems, in particular for the experiments.

In Ising-like chains (TMMC and the Co compounds), transitions from the groundstate to the soliton state are forbidden (see the dash line in Fig. 4a). In such systems, the presence of solitons is detected by looking at the low-energy fluctuations induced by the soliton motion \[\square\]. In Cu Benzoate, however, the soliton dispersion can be determined experimentally: the transitions from the groundstate to both the soliton and the breather branches are allowed, (see the arrows in Fig. 9). With Cu Benzoate, the discretization of the breather spectrum in a sine-Gordon model is now well established.

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FIG. 11. Field dependence of the first breather modes observed by ESR (Faraday configuration) in Cu Benzoate [3].

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