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Semi-phenomenological analysis of neutron scattering results for quasi-two dimensional quantum anti-ferromagnet

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ABSTRACT

The available results from the inelastic neutron scattering experiment performed on the quasi-two dimensional spin $\frac{1}{2}$ anti-ferromagnetic material La_2CuO_4 have been analysed theoretically. The formalism of ours is based on a semi-classical like treatment involving a model of an ideal gas of mobile vortices and anti-vortices built on the background of the Néel state, using the bipartite classical spin configuration corresponding to an XY-anisotropic Heisenberg anti-ferromagnet on a square lattice. The results for the integrated intensities for our spin $\frac{1}{2}$ model corresponding to different temperatures, show occurrence of vigorous unphysical oscillations, when convoluted with a realistic spectral window function. These results indicate failure of the conventional semi-classical theoretical model of ideal vortex/anti-vortex gas arising in the Berezinskii–Kosterlitz–Thouless theory for the low spin magnetic systems. A full fledged quantum mechanical formalism and calculations seem crucial for the understanding of topological excitations in such low spin systems. Furthermore, a severe disagreement is found to occur at finite values of energy transfer between the integrated intensities obtained theoretically from the conventional formalism and those obtained experimentally. This further suggests strongly that the full quantum treatment should also incorporate the interaction between the fragile-magnons and the topological excitations. This is quite plausible in view of the recent work establishing such a process in XXZ quantum ferromagnet on 2D lattice. The high spin XXZ quasi-two dimensional antiferromagnet like $MnPS_3$ however follows the conventional theory quite well.

1. Introduction

Spin dynamics in low dimensional magnetic systems have generated a significant research interest during the last three decades [1–31]. Different types of one and quasi-one dimensional as well as two and quasi-two dimensional systems have been studied both experimentally and theoretically to probe and understand the spin dynamics arising from the conventional spin wave excitations and their mutual interactions, as well as from vortex/meron and soliton like topological excitations [15–17,27,19,20]. In many of these systems the topological excitations of soliton and vortex/meron type occur naturally as they are thermodynamically feasible.

Motivated by the distinct possibilities of applications towards building of magnetic devices, the quasi-two dimensional systems have attracted a renewed research interest in recent times. Magnetic vortices present in these systems, have proved to be potential candidates for switching devices [32–35]. Direct experimental evidences of such vortices have been verified by both the Magnetic Force Microscopy (MFM) and the spin-polarized Scanning Tunnelling Microscopy (STM) [36,37].

The spin dynamics in many of the above magnetic systems has been investigated experimentally using the Inelastic Neutron Scattering (INS) and the Nuclear Magnetic Resonance (NMR) techniques. These include quasi-one dimensional systems such as $CsNiF_3$, layered systems such as K_2CuF_4, Rb_2CrCl_4, $LiCrO_2$, magnetically intercalated graphites, layered ruthenates, layered manganites and the high-T_C cuprates [16–18,20–29,31]. In the INS experiments performed on several of above materials, the existence of a prominent “central peak” (at $\hbar\omega = 0$) has been confirmed in the plot for the dynamical structure function $S(\mathbf{q}, \omega)$ vs. neutron energy transfer ‘$\hbar\omega$’ in the constant ‘$\mathbf{q}$’ scan [21,22]. These findings further serve as the motivation behind the huge variety of experiments performed on the layered magnetic systems. Moreover, the advancement of numerical and computational techniques also contributed to the understanding of the possible role of both the spin waves and the topological excitations in emergence of the central peak.

Kosterlitz and Thouless, and Berezinskii independently

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introduced the concept of vortex and anti-vortex like topological spin excitations in the two dimensional classical magnetic (spin) systems [38–40]. According to their ideas, there exists a non-conventional topological phase transition (known as Berezinskii–Kosterlitz–Thouless or BKT transition) characterized by the cross-over between binding to unbinding phases of vortex- anti-vortex pairs at a transition temperature T_{BKT}. Below this temperature all the vortices and anti-vortices are in a bound state and above this temperature some of them start moving freely. Further analytical and numerical studies and suitable extension of these approaches led to the proposal for the existence of topological vortices and anti-vortices in pure XY model and merons and anti-merons in XY-anisotropic Heisenberg model, for both ferromagnetic and anti-ferromagnetic types, on two-dimensional lattices [41–46]. Furthermore, approximate analytical calculations and Monte Carlo Molecular Dynamics (MCMD) simulations have strongly suggested that the freely moving topological excitations in the regime $T > T_{BKT}$, contribute non-trivially to the spin-spin correlation and give rise to the "central peak", as mentioned above [41–44]. The occurrence of such a "central peak" in quasi-two dimensional magnetic systems is now unanimously believed to be the signature for the dynamics of mobile topological excitations in a layer.

In spite of a lot of studies on the anisotropic Heisenberg model on two dimensional lattices the contributions of vortices/anti-vortices to the dynamics of the model, especially the detailed quantitative features of the DSFs are still not completely understood.

In the context of two-dimensional magnetism, the undoped (anti-ferromagnetic and non-superconducting) phases of the high T_c cuprate systems are believed to be excellent examples of two dimensional XY anisotropic Heisenberg Hamiltonian in an appropriate temperature regime. One member of this class of systems is La_2CuO_4, on which extensive INS experiments have been performed [30,31]. This is a truly spin-1/2 layered anti-ferromagnet. The intra-layer integrated intensity corresponding to the results of INS experiment performed on La_2CuO_4, exhibits a central peak when plotted against the neutron energy transfer $\hbar\omega$ (or frequency 'ω').

It has been shown that the results of vortex gas phenomenology and numerical simulations lead to an anomaly in the case of layered anti-ferromagnetic systems having very low spin values ($S=1/2$) [47]. Strikingly enough, the value of T_{BKT} obtained from Renormalization group analysis and numerical simulations is four (4) times the value of T_{BKT} calculated from the classical expression obtained by Kosterlitz and Thouless [47]. Furthermore, in our previous work we have already established that for quasi-two dimensional ferromagnetic systems having low spin values ($S=1/2$) the conventional semi-classical like treatment involving the ideal gas of unbound vortices/merons and anti-vortices/anti-merons corresponding to high temperature regime $T > T_{BKT}$, shows large inconsistency with the experimental situation and exhibits unphysical behaviour [48]. In this case the theoretical dynamical structure function (DSF) turns out to be negative for a wide range of energy transfers! However the range, over which the theoretical DSF remains positive, increases when the value of the spin is increased [48]. These facts motivate us to investigate and test in detail the applicability of the semi-classical-like treatment mentioned above to the INS results corresponding to real anti-ferromagnetic systems with $S=1/2$. For this exercise we select La_2CuO_4 as the reference system [30,31].

It is worthwhile to mention that the BKT transition can also be identified at the transition temperature T_{BKT} where the spin-stiffness jumps discontinuously from a universal value below T_{BKT} to zero above T_{BKT} [49]. Moreover, in anti-ferromagnetic model possessing Ising like anisotropy (in the z-direction) on two dimensional lattices, with external field being applied in the x-direction, such a discontinuous jump has been observed [50]. However, the discontinuous jump in this case may have its origin different from the vortex-anti-vortex unbinding mechanism since it is well known that the BKT transition in magnetic systems can only occur when the anisotropy is XY like [41–44].

The plan of the paper is as follows:- in Section 2 we describe the formulation of our semi-classical like treatment; in Section 3 we discuss our calculations and results and finally in Section 4 the conclusions and discussions of our present investigation are presented.

2. Mathematical formulation

The dynamics of mobile topological excitations in an anti-ferromagnetic system on a two-dimensional square lattice have been analysed both analytically and numerically [41–45]. The analytical studies have been performed by assuming a classical ideal gas of vortices/merons where the vortices/merons obey Maxwell's velocity distribution. The model system is described by the XY-anisotropic Heisenberg (XXZ) Hamiltonian, viz.,

$$\mathcal{H} = -J \sum_{\langle ij,pq \rangle} (S_{ij}^x S_{pq}^x + S_{ij}^y S_{pq}^y + \lambda S_{ij}^z S_{pq}^z), \tag{1}$$

where $\langle ij, pq \rangle$ label the nearest neighbour sites on a two-dimensional square lattice and $J(<0)$ is the anti-ferromagnetic exchange coupling. Here λ is the anisotropy parameter whose pure XY and isotropic Heisenberg limit correspond to $\lambda = 0$ and 1 respectively.

The structures of the vortices/merons have been obtained by solving the classical equations of motion corresponding to the Hamiltonian given by Eq. (1). In deriving the classical equations of motion the spins have been considered to be classical objects (classical spin fields $S(\mathbf{r}, t)$) as a function of position coordinates and time, which are defined on the entire lattice. At even or odd lattice sites these spin fields become identical to the following bipartite spin configurations:

$$\begin{aligned} S_{ij}^{even} &= +S\Big[\sin(\Theta_{ij} + \theta_{ij})\cos(\Phi_{ij} + \phi_{ij}), \sin(\Theta_{ij} + \theta_{ij})\sin(\Phi_{ij} + \phi_{ij}), \cos(\Theta_{ij} + \theta_{ij}))\Big], \\ S_{ij}^{odd} &= -S\Big[\sin(\Theta_{ij} - \theta_{ij})\cos(\Phi_{ij} - \phi_{ij}), \sin(\Theta_{ij} - \theta_{ij})\sin(\Phi_{ij} - \phi_{ij}), \cos(\Theta_{ij} - \theta_{ij}))\Big], \end{aligned} \tag{2}$$

where '*even*' and '*odd*' signifies the two different sub-lattices [51]. The static spin configuration corresponding to the merons are described by the capital angles $\Theta(\mathbf{r})$ (polar) and $\Phi(\mathbf{r})$ (azimuthal), and the time dependent small angles $\theta(\mathbf{r}, t)$ and $\phi(\mathbf{r}, t)$ describe the corresponding deviations from the static structure due to the motion of the merons and the spin dynamics above BKT transition temperature [44,45]. The expression of the vortex core radius is given by [44,45]

$$r_v = \frac{a}{2}\sqrt{\frac{\lambda}{1-\lambda}}. \tag{3}$$

From the above considerations the in-plane dynamical structure function (in-plane DSF) $S^{xx}(\mathbf{q}, \omega)$ is given by,

$$S^{xx}(\mathbf{q}, \omega) = \frac{S(S+1)}{2\pi} \frac{\gamma^3 \xi^2}{(\omega^2 + \gamma^2[1 + (\xi \mathbf{q}^*)^2])^2}, \tag{4}$$

with $\gamma = \frac{\sqrt{\pi}\bar{u}}{2\xi}$, where $\mathbf{q}^* = (\mathbf{q}_0 - \mathbf{q})$; $\mathbf{q}_0 = (\pi/a, \pi/a)$ and in our case $S = \frac{1}{2}$. The above expression for the in-plane dynamical structure function is a squared Lorentzian exhibiting a central peak at $\omega = 0$ in 'ω'-space for constant $\mathbf{q}$- scan and exhibiting a central peak at the zone boundary of the first Brillouin Zone (BZ) in the 'q' space for constant ω-scan [44,45]. In the above expression $\bar{u}$ is the root mean square (rms) velocity of the vortices and is given by,