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AUTHOR(S): F. G. Mertens
A. R. Bishop
G. M. Wysin
C. Kawabata

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DYNAMICAL CORRELATIONS FROM MOBILE VORTICES IN TWO-DIMENSIONAL EASY-PLANE FERROMAGNETS

F. G. Mertens†, A. R. Bishop, G. M. Wysin
Theoretical Division and Center for Nonlinear Studies
Los Alamos National Laboratory
Los Alamos, NM 87545, USA
and
C. Kawabata
Okayama University Computer Center
Okayama, 700, Japan

Abstract

Assuming an ideal gas of unbound vortices above the Kosterlitz-Thouless transition temperature, the dynamic form factors are calculated for both the in-plane and out-of-plane correlations. In both cases central peaks are predicted which are, however, produced by quite different mechanisms, depending on whether the correlations are globally or locally sensitive to the presence of the vortices. For the in-plane correlations the wavevector dependencies of the width and intensity of the peaks are very well supported by the central peaks, which are observed in a combined Monte Carlo-molecular dynamics simulation of the XY-model. Therefore, the parameters of the theory (rms vortex velocity and mean vortex-vortex separation) can be fitted and turn out to agree rather well with independent theoretical estimates. Recent inelastic neutron scattering experiments on the in-plane correlations for $BaCo_2(AsO_4)_2$ and Rb_2CrCl_4 also show central peaks. Their temperature and wavevector dependencies are consistent with our results, but their widths are larger than the theoretical estimates. Therefore, these peaks are interpreted to result, as least partially, from a gas of vortices. For the out-of-plane correlations our simulations also show a central peak. However, so far it cannot be identified unequivocally as a vortex contribution.

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† Permanent address: Physics Institute, University of Bayreuth, D-8580 Bayreuth, Fed. Rep. of Germany

1. Introduction

The increasing emergence of examples of well-characterized quasi-two-dimensional (2-D) magnetic materials has been prompted by technical advances in artificially structured, layered and surface-layer materials. More recently there has also been renewed attention to accurate inelastic neutron scattering measurements at low frequencies and long wavelengths in quasi 2-D magnets. It is therefore an appropriate time for detailed studies of 2-D spin dynamics. One particularly interesting case concerns materials with easy-plane symmetry, where we can probe dynamics associated with strongly nonlinear collective structures such as vortices and domains. For example, in pure easy-plane symmetry we expect that a Kosterlitz-Thouless[1] (K-T) type of topological phase transition will occur, with vortex-antivortex pairs beginning to unbind above a critical temperature T_c. In this regime it is then natural to ask whether there are dynamical signatures of the low density of unbound vortices. This is the principal concern of the present work.

Candidate materials are increasing rapidly and include: K_2CuF_4, Rb_2CrCl_4, $BaM_2(XO_4)_2$ (M = Co, Ni, ...; X = As, P, ...) and other layered magnets;[2-5] magnetically-intercalated graphites, e.g. $CoCl_2$-GIC prepared with various stagings;[6] and magnetic surface layers (e.g. magnetic lipids or magnetic epitaxial layers).[7] Treated within localized (Heisenberg) spin models (below), the ratio of inter- to intra-plane magnetic coupling constant is typically 10^{-3} - 10^{-6}. Furthermore, a great variety of magnetic interactions can be tuned by varying the material -- from ferromagnetic, to antiferromagnetic (e.g. $BaNi_2(PO_4)_2$), to competing nearest and next-nearest neighbors (e.g. $BaCo_2(AsO_4)_2$. These can have various degrees of (crystal field) symmetry-breaking in the easy plane, leading to domain patterns which compete with the characteristic vortex structures of the easy-plane symmetry.

Clearly this field is very rich in terms of materials and raises some fundamental questions with regard to <u>nonlinear</u> spin dynamics -- in much the same way that quasi-1-D magnets have challenged theoretical frameworks in the last decade.[8,9] Although dynamics associated with K-T theory has been studied successfully in the topologically equivalent problems of 2-D superfluids,[10] superconducting granular films,[11] and 2-D Josephson junction arrays,[12] comparable studies have not been made for 2-D magnets, except for some renormalized spin-wave approaches[13,14] and partial vortex-spinwave "phenomenologies" (below).

Since the scenario of vortex-antivortex pair unbinding introduced by K-T has been so successful for thermodynamic properties, it is important to test its predictive power for <u>dynamics</u>. Therefore, we will focus here on the phenomenology of an ideal, dilute gas of free vortices above T_c moving in the presence of renormalized spin waves and screened by the remaining vortex-antivortex bound pairs. Such an approach, explicitly incorporating the nonlinear coherent excitations, is similar in spirit to "soliton-gas" approaches for 1-D magnets[8] and has already been advocated by Huber.[15] However, he calculated only vortex autocorrelation functions, leading to dynamic form factors without any wave-vector dependence. Here we will calculate full form factors $S(\vec{q},\omega)$, and compare them with recent simulations[16] (Monte Carlo-molecular dynamics (MC-MD)) as well as inelastic neutron scattering data[17,18,5] -- both simulations and experiments have found anomalous "central peak" structures (i.e. scattering intensity near $\omega = 0$) for $T > T_c$, and indeed scattering from a vortex gas will be identified here as one mechanism for such a central peak.

In this paper we consider only the simplest situation of pure easy-plane spin symmetry. (Results for more general cases will be presented elsewhere.) However, dynamics necessarily involves some out-of-plane spin motion. Therefore, we treat explicitly the anisotropic Heisenberg model with classical Hamiltonian

$$H = -J \sum_{(m,n)} [S_x^m S_x^n + S_y^m S_y^n + \lambda S_z^m S_z^n] \quad , \tag{1.1}$$

where (m,n) label near-neighbor sites on a 2-D square lattice, J is a ferromagnetic coupling constant, and the classical spin vector is $\vec{S}^m = (S_x^m, S_y^m, S_z^m)$. The XY and isotropic Heisenberg limits correspond to $\lambda = 0$ and 1, respectively. Note that $\lambda = 0$ does not correspond to the "planar" limit[1] where spins are strictly confined to the XY plane. However, critical properties for the in-plane spin components (S_x or S_y) are still those of K-T theory -- e.g. with static spin-spin correlations changing from exponential to power law as T is decreased below T_c. By contrast the static correlations for the out-of-plane component (S_z) are exponential both above and below T_c (possibly with higher order signatures at T_c and the specific heat maximum at $T_s > T_c$).[19] The variation of T_c with λ is experimentally important. Both vortex theory[21] and MC results[20] show that T_c is only weakly-dependent on λ except for λ very close to 1, when $T_c \to 0$. Thus, even materials with very weak easy-plane anisotropy (e.g., $\lambda \simeq 0.99$ in K_2CuF_4) have a substantial K-T transition temperature and 2-D fluctuation regime -- the true ordering, sufficiently close to the actual transition, is of course 3-D in real materials. For the materials mentioned above, coupling constants have been estimated from fits to, e.g. linear spin wave theory, and λ values are in the range 0.4 - 0.99, where T_c^{2-D} is still close to $T_c^{2-D}(\lambda = 0)$.

We will need to make use later of thermodynamic results of K-T theory. Assuming we can adopt planar limit results as a guide, the relevant information for our purposes primarily concerns the static correlations[22-25]

$$S_{xx}(r) \sim r^{-1/2} \exp(-r/\xi(T)) \quad , \quad T > T_c \tag{1.2}$$

where ξ is the correlation length and

$$\xi(T) = \xi_0 \exp(b\tau^{-1/2}) \quad , \quad \tau \equiv (T-T_c)/T_c \quad . \tag{1.3}$$

ξ_0 is on the order of the lattice constant, and b has been found[25] to be quite temperature-dependent even for small τ. The correlation length can be further interpreted[10] as half of the mean separation between free vortices:

$$n_v(\tau) \simeq (2\xi)^{-2} \quad , \tag{1.4}$$

with n_v the free vortex density. Of course, these should only be viewed as order-of-magnitude relations, since the vortex creation energy itself decreases as λ increases,[21] and full thermodynamics of the anisotropic Heisenberg model (1.1) are not available. Although such effects can be partially included, we prefer to leave form (1.3) and compare it with fits of ξ to our numerical data (below). Ultimately a direct estimate of n_v from numerical simulations (following individual vortex dynamics) may itself be possible, cf. ref. 26.

As in 1-D easy-plane magnets, careful distinction must be made between in-plane and out-of-plane dynamic correlations. In addition to the remarks concerning critical properties above, we will find in section 2 that a central peak for $S_{xx}(\vec{q},\omega)$ is predicted to arise above T_c

from a vortex gas. The correlations reveal the mean vortex-vortex separation 2ξ and the rms vortex velocity $\bar{u}$. These phenomenological parameters are determined by fitting the width and intensity of the predicted central peak to the corresponding quantities from our MC-MD simulations. The results are compared with theoretical estimates: ξ from (1.3) and $\bar{u}$ from the velocity autocorrelation function of Huber.[15] (In our publication[40] of preliminary results we did not fit the parameters but rather used the theoretical estimates.)

Besides the central peaks, our MC-MD results also show spin-wave contributions. Above T_c these are strongly softened for S_{xx} (consistent with the "universal jump" prediction[27]), but not for S_{zz}. Moreover, there seem to be multi-magnon contributions, especially for S_{zz}.

In section 3 our results are compared with recent inelastic neutron scattering experiments. So far central peaks in $S_{xx}(\vec{q},\omega)$ have been reported for Rb_2CrCl_4 (ref. 17) and $BaCo_2(AsO_4)_2$ (refs. 18, 5). We discuss in detail the ω-, q-, and temperature dependencies of the peaks.

In section 4, $S_{zz}(\vec{q},\omega)$ is calculated assuming that the out-of-plane structure of moving vortices can be approximated by the static structure. The width of the predicted central peak is consistent with our MC-MD results, but not the intensity. We conclude that the velocity dependence of the out-of-plane structure must be incorporated.

Section 5 contains a summary and a discussion of the limitations of the present theory, and of possible modifications which take into account the great variety of different interactions and symmetries of the real materials.

2. In-Plane Correlations

We use a continuum description and spherical coordinates for a general time-dependent spin configuration:

$$S_x(\vec{r},t) = S \cos\phi(\vec{r},t) \sin\theta(\vec{r},t)$$

$$S_z(\vec{r},t) = S \cos\theta(\vec{r},t) \tag{2.1}$$

with $\vec{r} = (x,y)$. Consider a single vortex at the origin:

$$\phi(\vec{r}) = \pm \tan^{-1}(y/x) \quad . \tag{2.2}$$

The form of $\theta(\vec{r})$ will be discussed in section 4. The vortex solutions have the asymptotic properties

$$\theta_{\pm}(r) = \begin{cases} \frac{\pi}{2}\,[1 \mp \exp(-r/r_v)] \quad , \; r \gg r_v & (2.3a) \\ 0 \text{ or } \pi \quad , \; r \to 0 & (2.3b) \end{cases}$$

with vortex-core "radius" r_v. S_z is localized and correlations are sensitive to the vortex size and shape (section 4). By contrast, S_x (and S_y) are not localized, i.e. they have no spatial Fourier transform. Therefore the in-plane correlation function $S_{xx}(\vec{r},t) = \langle S_x(\vec{r},t)\, S_x(\vec{0},0)\rangle$ is only globally sensitive to the presence of vortices, which act to break long-range order in $\cos\phi$. Thus the characteristic length is the mean vortex-vortex separation 2ξ.

Consider first the field $\cos\phi(\vec{r},t)$ in $S_x = S\cos\phi\sin\theta$. As seen in Fig. 1 for a particular case, every vortex that passes with its center between $\vec{0}$ and $\vec{r}$ in time t diminishes the correlations, changing $\cos\phi$ by a factor of (-1), independent of the direction of movement. In this sense vortices act like "2-D sign functions." Inclusion of the field $\sin \theta(\vec{r},t)$ in S_x, with θ given by (2.3), shows that the change of sign does not occur abruptly, but over the distance $2r_v$. This means that vortices behave effectively as "2-D kinks" with half width r_v. Considering length scales $\gg r_v$ the dominant effect of the moving vortices are the above-mentioned changes of sign. Thus an ideal vortex gas gives

$$S_{xx}(\vec{r},t) = S^2 \langle \cos^2\phi \rangle \langle (-1)^{N(\vec{r},t)} \rangle \quad . \tag{2.4}$$

Here $N(\vec{r},t)$ is the number of vortices which pass an arbitrary, non-intersecting contour connecting $(\vec{0},0)$ and $(\vec{r},t)$; the average over $\cos^2\phi$ is 1/2, assuming a random spin configuration outside of the vortex cores.

Expressions like (2.4) were evaluated by several authors for the case of kinks in 1-D models (e.g. ϕ^4 or sine-Gordon), see ref. 8 and references cited therein. A very detailed investigation was made by Dorogovtsev,[29] who also calculated such correlations numerically in two dimensions. We have adopted his general procedure and, by implementing several modifications, we identify certain cancellations which allow us to calculate (2.4) analytically. We will demonstrate this for the 1-D case, the generalization to higher dimensions will then be straightforward.

For simplicity all kinks are first taken to have the same velocity u. Considering the case $x \geq ut > 0$, we choose the velocity-independent contour $(0,0) \rightarrow (x,0) \rightarrow (x,t)$, which is outside of the "light" cone $x = \pm ut$. The contribution from the first part of this contour is

$$<(-1)^{N(x,t)}> = \sum_{n_\ell} (-1)^{n_\ell} p(n_\ell) \sum_{n_r} (-1)^{n_r} p(n_r) \quad . \tag{2.5}$$

Because we assumed a <u>dilute</u> ideal gas, we have a Poisson distribution, $p(n_\ell) = \bar{n}_\ell^{\,n_\ell}/n_\ell! \exp(-\bar{n}_\ell)$, where n_ℓ and n_r are the numbers of kinks in $[0,x]$ running to left and to right, respectively; $\bar{n}_\ell$ and $\bar{n}_r$ are the average numbers. Kinks pass this part of the contour at the same time (t=0) but at different positions, implying that these events are not correlated. Thus n_ℓ and n_r are independent, and the two sums in (2.5) can be calculated separately, giving $\exp(-2\bar{n}_\ell - 2\bar{n}_r) = \exp(-x/\xi)$, for (2.5). For the second part of the contour, $(x,0) \rightarrow (x,t)$, we obtain formally the same expression as (2.5), but with n_ℓ the number of left-running kinks in $[x, x + ut]$, and n_r the number of right-running kinks in $[x - ut,x]$. Kinks pass the same point at different times. Events are <u>correlated</u> as long as $t < x/u$, which is just the case we are considering. Thus n_ℓ and n_r are <u>not independent</u>. Assuming $n_\ell = n_r$, then $(n_\ell + n_r)$ is even, implying that there is no contribution from the second part of the contour.

Now consider the second case, $ut \geq x > 0$. We choose the contour $(0,0) \rightarrow (0,t) \rightarrow (x,t)$, inside the light cone. The same type of argument shows that the vertical component $(0,0) \rightarrow (0,t)$ gives a contribution, whereas the horizontal part $(0,t) \rightarrow (x,t)$ does not -- precisely opposite to the first case. The result for (2.5) is $\exp(-ut/\xi)$, in contrast to $\exp(-x/\xi)$ in the first case. Both cases can be combined into the final result,

$$<(-1)^{N(x,t)}> = \exp \{- \frac{|x-ut|}{2\xi} - \frac{|x+ut|}{2\xi}\} \quad . \tag{2.6a}$$

The 2-D calculation, where the vortices play the role of the kinks, proceeds in a similar way (Appendix B). Including an average over all velocities $u = |\vec{u}|$, we have

$$<(-1)^{N(\vec{r},t)}> = \exp \{ - \int_0^\infty [\frac{|r-ut|}{2\xi} + \frac{|r+ut|}{2\xi}] \tilde{P}(u)\, du\} \quad . \tag{2.6b}$$

In order to obtain the velocity distribution we consider the velocity $\vec{u}$ of a vortex at $\vec{R}$ which results from an equation of motion.[15] $\vec{u}$ is proportional to $\hat{z} \times \vec{F}$; here $\hat{z}$ is a unit vector perpendicular to the XY-plane and $\vec{F}$ is the net force due to the interactions with the other vortices at $\vec{R}_\nu$. These forces are proportional to $\vec{R} - \vec{R}_\nu$ and decrease with increasing distance. A perfectly symmetric array of the $\vec{R}_\nu$ would yield a zero net force and thus zero velocity. However, the density of vortices is homogeneous only on the average, locally the distribution is random. Therefore, the deviations from the average $\vec{u} = 0$ follow a Gaussian normal distribution, i.e. we can assume a Maxwellian velocity distribution $P(\vec{u})$. For the distribution $\tilde{P}(u)$ of the moduli we get $\tilde{P}(u) \sim u$ times a Gaussian (Appendix B). The integration over u in (2.6b) eventually leads to

$$S_{xx}(\vec{r},t) = \frac{1}{2} S^2 \exp \{-\frac{r}{\xi} - \frac{\sqrt{\pi}}{2} \frac{\bar{u}|t|}{\xi} \operatorname{erfc} (\frac{r}{\bar{u}t})\} \quad , \tag{2.7}$$

where $\bar{u}$ is the root-mean-square velocity and erfc is the complementary error function. Similarly to the 1-D case[30], there is an excellent analytic approximation for (2.7), which preserves not only the integrated intensity (see below) but also the correct asymptotic behavior for r or t → ∞. Namely,

$$S_{xx}(\vec{r},t) \simeq \frac{1}{2} S^2 \exp \{-[(r/\xi)^2 + (\gamma t)^2]^{\frac{1}{2}}\} \quad , \tag{2.8}$$

with $\gamma = \sqrt{\pi}\ \bar{u}/(2\xi)$. Using approximation (2.8), both the spatial and temporal Fourier transforms can be performed, yielding (Appendix C)

$$S_{xx}(\vec{q},\omega) = \frac{S^2}{2\pi^2} \frac{\gamma^3\xi^2}{\{\omega^2 + \gamma^2[1 + (\xi q)^2]\}^2} \quad . \tag{2.9}$$

This is a (squared) Lorentzian central peak (c.p.) with q-dependent width

$$\Gamma_x(q) = \frac{1}{2} [\pi(\sqrt{2} - 1)]^{\frac{1}{2}} \frac{\bar{u}}{\xi} \sqrt{1 + (\xi q)^2} \quad , \tag{2.10}$$

and integrated intensity

$$I_x(q) = \frac{S^2}{4\pi} \frac{\xi^2}{[1 + (\xi q)^2]^{3/2}} \quad . \tag{2.11}$$

(Result (2.11) can be checked by performing the Fourier transform of (2.7) with t = 0).

Note that $I_x \sim \xi^2 \sim n_v^{-1}$, as expected since the correlations are diminished by the presence of the vortices. (In contrast to the out-of-plane correlations where $I_z \sim n_v$, see section 4).

We now compare the predictions of this phenomenological theory with the results of our MC-MD simulation. For these simulations we have used Hamiltonian (Landau) spin dynamics $d\vec{S}_n/dt = \{\vec{S}_n,H\}$ with Hamiltonian

(1.1) on an isotropic square lattice with a size up to 100 × 100, giving accurate access to wave numbers $\gtrsim 0.02\ \pi/a$. The MC-algorithm[19] used 10^4 MCS per spin to equilibrate 3 random initial configurations. Then MD with 4th-order Runge-Kutta was applied, with timestep 0.04, sampling time NS × 0.04, NS = 4, 8, 32, and total integration time 512 × NS × 0.04. This is in units where $J = k_B = a = S = 1$. Further details are given in Appendix D.

Figure 2 shows $S_{xx}(\vec{q},\omega)$ from our MC-MD simulations for the XY-model. The spin waves are strongly softened for $T > T_c$, consistent with the theoretical predictions[27] of a "universal jump" as well as with experiments.[2,17,18,5] However, the spin-wave softening depends[14] on q, i.e. on the length scale over which vortices are considered to be free (c.f. the analogous situation in 2-D superfluids[10] and 2-D Josephson junction arrays[12]). Therefore we observe for small q only a central peak; for large q broad spin-wave contributions can be distinguished besides the c.p.

The predicted q-dependence (2.10) of the c.p. width is very well supported by the MD data (Fig. 3). For $q \gg \xi^{-1}$, we have $\Gamma_x = 0.57\ \bar{u}q$, and a fit of $\bar{u}$ gives the numbers in Table 1; they can be compared with a formula of Huber[15] obtained from the velocity autocorrelation function using an equation of motion for free vortices:

$$\bar{u}^2 = \frac{\pi}{2} \left(\frac{JS^2a^2}{\hbar}\right)^2 n_v \ln\left(\frac{k_B T_c}{JS^2 n_v a^2}\right) . \tag{2.12}$$

The logarithmic term can be approximated using (1.3), (1.4), and $\xi_0 = a$. With $T_c \simeq 0.8$ (ref. 19) we get