Vortex Signatures in Dynamic Structure Factors for Two-Dimensional Easy-Plane Ferromagnets

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(Received 5 February 1987)

The *XY* and the anisotropic Heisenberg models are considered above the Kosterlitz-Thouless transition temperature. Under the assumption of a gas of freely moving vortices, it is shown that the dynamic structure factor exhibits a central peak for both in-plane and out-of-plane correlations, in good agreement with the results of a combined Monte Carlo–molecular-dynamics simulation. These results are also consistent with recent neutron-scattering data on Rb_2CrCl_4 and $BaCo_2(AsO_4)_2$, which show qualitatively the same wave-vector and temperature dependencies.

PACS numbers: 75.25.+z, 05.20.Dd

The Kosterlitz-Thouless (KT) theory[1] of topological phase transitions in two spatial dimensions (2D) has found many successful applications.[2] However, the phenomenological scenario of vortex-antivortex pairs unbinding above a transition temperature T_c has been difficult to probe *dynamically*—with the important exceptions of 2D superfluids[3] and superconducting granular films.[4] The emergence[5-7] of well-characterized quasi 2D easy-plane magnetic materials and relevant inelastic neutron scattering opens the way to studying dynamic signatures of nonlinear spin excitations in 2D, including vortices.

As a first step, we have considered quasi 2D Heisenberg ferromagnets with easy-plane anisotropy. The opportunity here is comparable to that exploited recently in quasi 1D easy-plane magnets[8,9] and we have adopted a similar philosophy—extensive Monte Carlo–molecular-dynamics (MC-MD) simulations, and comparisons with a phenomenology of "ideal gases" of unbound vortices[10] and spin-waves (above T_c), and with experimental data. According to KT theory the unbound vortices above T_c move in a screening background of the remaining bound pairs; such effects are grossly incorporated via equilibrium-thermodynamic input.[11] For simplicity we have assumed Hamiltonian (Landau) spin dynamics $d\mathbf{S}_n/dt=\{\mathbf{S}_n,H\}$ (with spin $\mathbf{S}_n$ at site n). The MC-MD studies[12] were performed on isotropic square lattices with dimensions up to 100×100 giving accurate access to wave vectors $\gtrsim(0.02)\pi/a$. Previous studies[13] have demonstrated the *weak* sensitivity of T_c to the easy-plane symmetry-breaking strength, as well as interesting features in out-of-plane static correlations. Here also we find that dynamic signatures of spin waves and vortices carry quite distinct structure and information for in-plane and out-of-plane correlations.[8-10] Our major conclusions are the striking agreements between ideal-gas phenomenology and MC-MD simulations, and the strong qualitative similarities with available inelastic-neutron-scattering data.[5,6]

Specifically, we consider the anisotropic Heisenberg Hamiltonian[12,13]

$$H=-J\sum_{(m,n)}[S_x^mS_x^n+S_y^mS_y^n+\lambda S_z^mS_z^n], \quad (1)$$

where the nearest-neighbor pairs (m,n) span a 2D square lattice (x,y) and $0\le\lambda<1$. Continuum vortex spin configurations obey[14]

$$\phi=\tan^{-1}(y/x),$$
$$\theta=\begin{cases}\pi/2[1-e^{-r/r_v}], & r\gg r_v,\\ 0, & r\to0,\end{cases} \quad (2)$$

with $S_x=S\cos\phi\sin\theta$, $S_z=S\cos\theta$, $r^2=x^2+y^2$, and r_v a vortex core "radius" $a[2(1-\lambda)]^{-1/2}$ (lattice constant a). We find below that S_z is only locally sensitive to vortices, whereas S_x (or S_y) is globally sensitive. Thus, in-plane and out-of-plane correlations reveal mean vortex-vortex *separation* and vortex *shape*, respectively (cf. 1D[8,9]).

Out-of-plane correlations.—We approximate an arbitrary field configuration by a sum of spin-wave and vortex contributions. The vortex contribution is taken as an ideal gas of N_v free vortices with positions $\mathbf{R}_v$ and velocities $\mathbf{u}_v$:

$$S_z(\mathbf{r},t)\simeq S\sum_{v=1}^{N_v}\cos\theta(\mathbf{r}-\mathbf{R}_v-\mathbf{u}_vt). \quad (3)$$

The vortex dynamic correlation function $S_{zz}(\mathbf{r},t)=\langle S_z(\mathbf{r},t)S_z(\mathbf{0},0)\rangle$ is evaluated[8] with incoherent scattering from the independent vortices, assuming a Maxwellian distribution of $\{\mathbf{u}_v\}$. Transforming in $\mathbf{r}$ and t gives

$$S_{zz}(\mathbf{q},\omega)=\frac{S^2}{4\pi^{5/2}}\frac{n_v}{\bar{u}}\frac{|f(q)|^2}{q}e^{-\omega^2/(\bar{u}q)^2}, \quad (4)$$

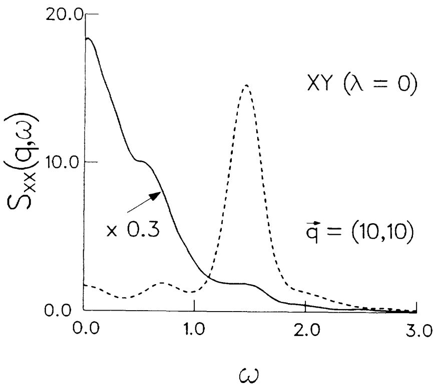

FIG. 1. Smoothed dynamic structure factor for *out-of-plane* correlations from MC-MD; $\mathbf{q}$ in units of $2\pi/L$, with lattice size $L=100a$. Temperature $T=0.5$ (dashed line) and 1.1 (solid line), with $T_c \simeq 0.83$.

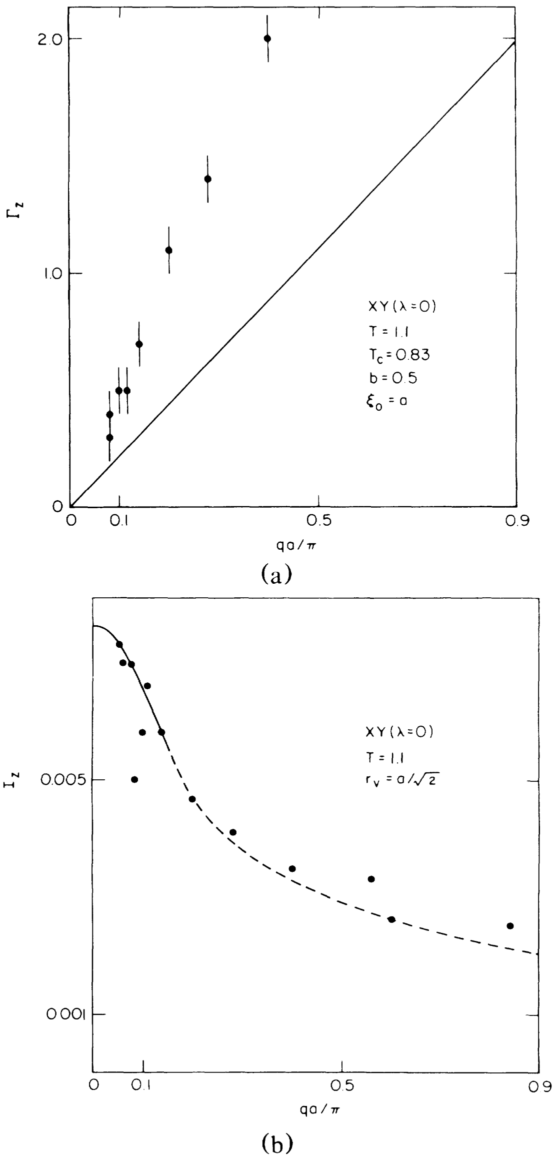

FIG. 2. Width Γ_z and intensity I_z of S_{zz} central peak. Data points and error bars result from estimating Γ_z and I_z from plots like Fig. 1. Solid lines result from the Gaussian (4), without parameter fitting; dashed line in (b) is a guide to the eye.

with n_v the vortex density and $\bar{u}$ the rms speed. The vortex form factor $f(q)$ [the Fourier transform of $\cos\theta(\mathbf{r})$] is evaluated approximately by extending (2) to small r and expanding about $\theta=\pi/2$: In first order this gives

$$f(q) \simeq \pi^2 r_v^2 [1+(qr_v)^2]^{-3/2}, \quad qr_v \ll 1. \qquad (5)$$

From studies of *XY* model *thermodynamics*,[1-4,11] we expect $n_v(T)=\xi^{-2}(T)$, with correlation length $\xi=\xi_0 \times \exp(b\tau^{-1/2})$, $\tau=(T-T_c)/T_c$, $\xi_0=O(a)$, and $b \approx 0.3$–0.5 for temperatures considered below. Huber[10] has calculated $\bar{u}(T)=(\pi b)^{1/2} JS^2 a^2 \hbar^{-1} n_v^{1/2} \tau^{-1/4}$ (in the absence of dissipation).

Figures 1–4 compare ideal-gas predictions with our MC-MD simulation results for the *XY* limit ($\lambda=0$).[15] Below T_c (≈ 0.83 in units of J/k_B), there is only a spin-wave component. This is not strongly affected for $T>T_c$ but an *additional central peak* (CP) appears (Fig. 1; $T=1.1$). From (4), the CP *width* Γ_z is predicted as $\bar{u}q$. This linear form is well supported by the MC-MD data [Fig. 2(a)]—the observed slope is greater by a factor of ~ 2; however, width estimates from the data are upper bounds and theoretical estimates of b are very approximate. We could fit the slope with another ξ_0 (for which only the order of magnitude is known). Interestingly, we predict Γ_z to *saturate* as $\tau \gtrsim 0.5-$ (for $b=0.5$), and we observe a nearly constant Γ_z for $\tau \gtrsim 0.1$. The CP integrated *intensity* I_z is predicted from (4) as $I_z(q)=n_v S^2 (2\pi)^{-2} |f(q)|^2$. Using the quantum theory value[14] $r_v=a/\sqrt{2}$, we find good agreement with MC-MD data for $q \lesssim (2r_v)^{-1}$ [Fig. 2(b)]. This agreement is not expected for larger q since we approximated $\theta(r)$ for small r. Note that $r_v \simeq 0.7a$ implies that spins are strongly constrained to the *XY* plane even near the vortex core—consistent with our simulations. The observed absolute values of I_z are an order of magnitude smaller than predicted, probably because of destructive interference with magnons.[16] The predictions that Γ_z saturates at finite τ and $I_z \propto n_v$ differ from those of Ref. 10, where $\Gamma_z \propto n_v$, $I_z \propto n_v^2$.

In-plane correlations.—Correlations of $S_x(\mathbf{r},t)$ with $S_x(\mathbf{0},0)$ are globally sensitive to vortices. All vortices with centers passing between $\mathbf{0}$ and $\mathbf{r}$ in time t diminish the correlations, changing $\cos\phi$ by $\sim(-1)$ (except for a measure zero set moving along the x or y axes): vortices act like 2D sign functions. Considering length scales $\gg r_v$, we assume the ideal-vortex-gas form[17]: $S_{xx}(\mathbf{r},t)$

$=S^2\langle\cos^2\phi\rangle\langle(-1)^{N(\mathbf{r},t)}\rangle$, where $N(\mathbf{r},t)$ is the number of vortices passing an arbitrary, nonintersecting contour connecting $(\mathbf{0},0)$ and $(\mathbf{r},t)$.[18] In the spirit of Ref. 18, we use a velocity-independent contour $(\mathbf{0},0)\rightarrow(\mathbf{r},0)\rightarrow(\mathbf{r},t)$ and make use of various cancellations (depending on whether or not part of the contour is in the "light" cone $r=|\mathbf{u}|t$). Assuming again a Maxwellian velocity distribution, we find

$$S_{xx}(\mathbf{r},t)=\tfrac{1}{2}S^2\exp-[r/\xi+\tfrac{1}{2}\pi^{1/2}(\bar{u}|t|/\xi)\mathrm{erfc}(r/\bar{u}t)]. \tag{6}$$

An excellent analytic approximation for the argument of the exponential in (6) is $\{(r/\xi)^2+(\gamma t)^2\}^{1/2}$, where $\gamma=\frac{1}{2}\pi^{1/2}\bar{u}/\xi$ (cf. Takayama and Maki[19]). This approximation preserves the correct asymptotic behaviors as $|t|$ or $r\rightarrow\infty$, and also the integrated intensity $I_x=(S^2/4\pi)\xi^2[1+(\xi q)^2]^{-3/2}$. The approximate dynamic structure factor is

$$S_{xx}(\mathbf{q},\omega)=\frac{S^2}{2\pi^2}\frac{\gamma^3\xi^2}{\{\omega^2+\gamma^2[1+(\xi q)^2]\}^2}. \tag{7}$$

Comparing (4) and (7), note the characteristic length scales r_v and ξ for S_{zz} and S_{xx}, respectively, and the Gaussian versus (squared) Lorentzian CP shapes.

Comparisons of (7) with our MC-MD data are again extremely good. Contrary to S_{zz}, the spin-wave peaks are strongly softened,[20] producing a central peak (Fig. 3): A proportionality to $[1+(\xi q)^2]^{1/2}$ is indeed observed for its width Γ_x [Fig. 4(a)], with good quantitative agreement using the theoretical estimates for $\bar{u}$ and ξ (from $b=0.5$, $\xi_0=a$).[11] Further, Γ_x is predicted to increase with τ and saturate at $\tau\simeq0.5$ for $q\xi\gg1$ and at high τ for $q\xi\ll1$. These behaviors are observed. The temperature dependence of the intensity I_x is governed by n_v^{-1}. Using the theoretical prediction for ξ, we find good agreement [Fig. 4(b)] with the simulations for $I_x(q)$, with $q\lesssim\xi^{-1}$. (Our approximations are best for large r.) The absolute values of I_x are about a factor of 5 larger than observed.[17]

Experimental inelastic-neutron-scattering results on XY-like magnets are presently incomplete. However, certain encouraging comparisons are worth remarking. The materials $BaCo_2(AsO_4)_2$ (Ref. 5) and Rb_2CrCl_4 (Ref. 6) appear to be good candidates. (Other potential examples include[7] K_2CuF_4 and high-stage magnetically intercalated graphite.) There is qualitative agreement between the observed and predicted temperature dependence of Γ_x in both $BaCo_2(AsO_4)_2$ and Rb_2CrCl_4 and orders of magnitude are also consistent. For instance, in

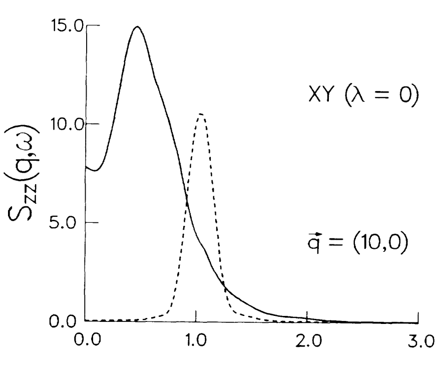

FIG. 3. Smoothed dynamic structure factor for *in-plane* correlations from MC-MD; details as in Fig. 1.

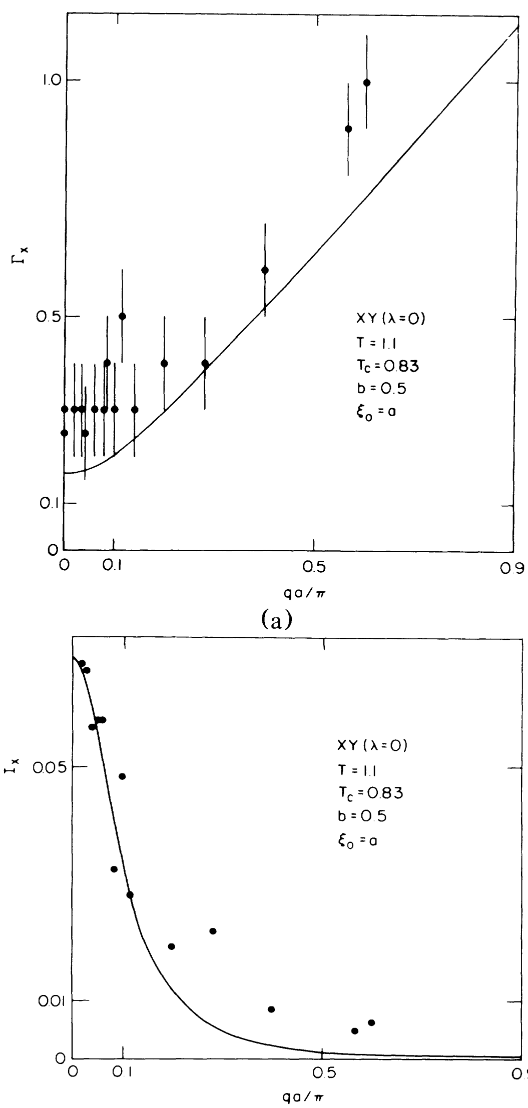

FIG. 4. Width Γ_x and intensity I_x of S_{xx} central peak. Data points and error bars result from estimating Γ_x and I_x from plots like Fig. 3. Solid lines results from the squared Lorentzian (7), without parameter fitting.

Rb_2CrCl_4, $\Gamma_x(q=0, \tau \simeq 0.015) \simeq 0.014$ meV, whereas (8) gives $\simeq 0.005$ and 0.05 meV for $b=1.5$ and 1.0, respectively. In addition, the reported q dependence of Γ_x for Rb_2CrCl_4 is in qualitative agreement with (7). It will be important to fit[6] existing and future experimental data with (7). Complementary measurements[6] of $S_{zz}(\mathbf{q},\omega)$, although difficult, are needed to isolate more clearly unbound vortex and spin-wave (and multiple-spin-wave) contributions.

In conclusion, our studies demonstrate the coexistence of spin-wave and vortex contributions to $S(\mathbf{q},\omega)$ above T_c in qualitative agreement with inelastic-neutron-scattering experiments: free vortices give rise to *central* ($\omega \approx 0$) scattering components of very different character for S_{xx} and S_{zz}; spin-wave softening occurs (at T_c) only for S_{xx}; ideal-gas phenomenology provides successful fitting forms. These results support the opportunities[5,13] for studying nonlinear excitations and dynamics in quasi 2D magnets more generally—including effects of in-plane crystalline fields and competing interactions,[5] which will provide additional low-frequency scattering from coherent structures. Future theoretical studies include vortex-vortex and vortex–spin-wave interactions and extrinsic dissipation (lifetime) mechanisms. In addition, several quasi 2D magnets are low-spin (e.g., K_2CuF_4 and $BaCo_2(AsO_4)_2$ are $S=\frac{1}{2}$). Thermodynamic studies suggest that the main quantum effects are substantial renormalizations (reductions) of intensities (of specific heat, etc.).[21,22] Describing quantum *dynamics* remains a major theoretical challenge in both 1D and 2D.

We are grateful for informative discussions with W. C. Kerr. The research of one of us (F.G.M.) was partially supported by a travel grant from the German Science Foundation.

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