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VORTEX SIGNATURES IN DYNAMIC STRUCTURE FACTORS FOR TWO-DIMENSIONAL EASY-PLANE FERROMAGNETS

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ABSTRACT

The XY- and the anisotropic-Heisenberg models are considered above the Kosterlitz-Thouless transition temperature. Assuming 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 wavevector and temperature dependencies.

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

As a first step, we have considered quasi-2-d Heisenberg ferromagnets with easy-plane anisotropy. The opportunity here is comparable to that exploited recently in quasi-1-d 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 K-T 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 $d\vec{S}_n/dt = \{\vec{S}_n, H\}$ (with spin $\vec{S}_n$ at site n). The MC-MD studies[12] were performed on isotropic square lattices with dimensions up to 100×100 giving accurate access to wavevectors $\gtrsim (0.02)\pi/a$. 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]

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

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

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

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" $a[2(1-\lambda)]^{-\frac{1}{2}}$ (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 <u>separation</u> and vortex <u>shape</u>, respectively (c.f. 1-d[8,9]).

<u>Out-of-plane correlations</u>. 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 $\vec{R}_\nu$ and velocities $\vec{u}_\nu$:

$$S_z(\vec{r},t) \simeq S \sum_{\nu=1}^{N_v} \cos\theta\,(\vec{r} - \vec{R}_\nu - \vec{u}_\nu t) \quad . \tag{3}$$

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

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

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

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

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

Figs. (1-4) compare ideal gas predictions with our MC-MD simulation results for the XY limit ($\lambda = 0$).[15] Below T_c($\simeq$ 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 (c.p.) appears (Fig. 1; T = 1.1). From (4), the c.p. 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 $\tau \to 0.5$- (for b = 0.5), and we observe a nearly constant Γ_z for $\tau \gtrsim 0.1$. The c.p. integrated <u>intensity</u> I_z is predicted from (4) as $I_z(q) = n_v S^2 (2\pi)^{-2} |f(q)|^2$. Using the continuum 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 $\Gamma_z \propto n_v$, $I_z \propto n_v^2$.

<u>In-plane correlations</u>. Correlations of $S_x(\vec{r},t)$ with $S_x(\vec{0},0)$ are globally sensitive to vortices. All vortices with centers passing between $\vec{0}$ and $\vec{r}$ in time t diminish the correlations, changing cos ϕ by ~(-1) (except for a measure zero set moving along the x- or y-axes): vortices act like 2-d sign functions. Considering length scales >> r_v, we assume the ideal vortex gas form[17]: $S_{xx}(\vec{r},t) = S^2 \langle\cos^2\phi\rangle \langle(-1)^{N(\vec{r},t)}\rangle$, where $N(\vec{r},t)$ is the number of vortices passing an arbitrary, non-intersecting contour connecting $(\vec{0},0)$ and $(\vec{r},t)$.[18] In the spirit of Ref. 18, we use a velocity-independent contour $(\vec{0},0) \rightarrow (\vec{r},0) \rightarrow (\vec{r},t)$ and make use of various cancellations (depending on whether or not part of the contour is in the "light"-cone $r=|\vec{u}|t$). Assuming again a Maxwellian velocity distribution, we find

$$S_{xx}(\vec{r},t) = \frac{1}{2}S^2 \exp - \{\frac{r}{\xi} + \frac{1}{2}\pi^{\frac{1}{2}}(\frac{\bar{u}|t|}{\xi})\mathrm{erfc}(\frac{r}{\bar{u}t})\} \quad . \tag{6}$$

An excellent analytic approximation for the argument of the exponential in (6) is $\{(r/\xi)^2 + (\gamma t)^2\}^{\frac{1}{2}}$, where $\gamma = \frac{1}{2}\pi^{\frac{1}{2}}\bar{u}/\xi$ [c.f. Ref.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

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

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 c.p. 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 $[1+(\xi q)^2]^{\frac{1}{2}}$ 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, ξ_0 = a).[11] Further, Γ_x is predicted to increase with τ and saturate at $\tau \simeq 0.5$ for $q\xi >> 1$ and at high τ for $q\xi << 1$. 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 $BaCo_2(As)_4)_2$ (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 Rb_2CrCl_4, Γ_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 to (7). Complementary measurements[6] of $S_{zz}(\vec{q},\omega)$, although difficult, are needed to isolate more clearly unbound vortex and spin-wave (and multi-spinwave) contributions.

In conclusion, our studies demonstrate the coexistence of spin-wave and vortex contributions to $S(\vec{q},\omega)$ above T_c in qualitative agreement with inelastic neutron scattering experiments: free vortices give rise to <u>central</u> ($\omega \simeq 0$) 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-2-d 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-spinwave interactions and extrinsic dissipation (lifetime) mechanisms. In addition, several quasi-2-d magnets are low-spin (e.g. K_2CuF_4 and $BaCo_2(As)_4)_2$ 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 <u>dynamics</u> remains a major theoretical challenge in both 1-d and 2-d.

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Figure Captions

Fig. 1. Smoothed dynamic structure factor for out-of-plane correlations from MC-MD; $\vec{q}$ in units of $2\pi/L$, with lattice size L = 100 a. Temperature T = 0.5 (---) and 1.1 (——), 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.

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.

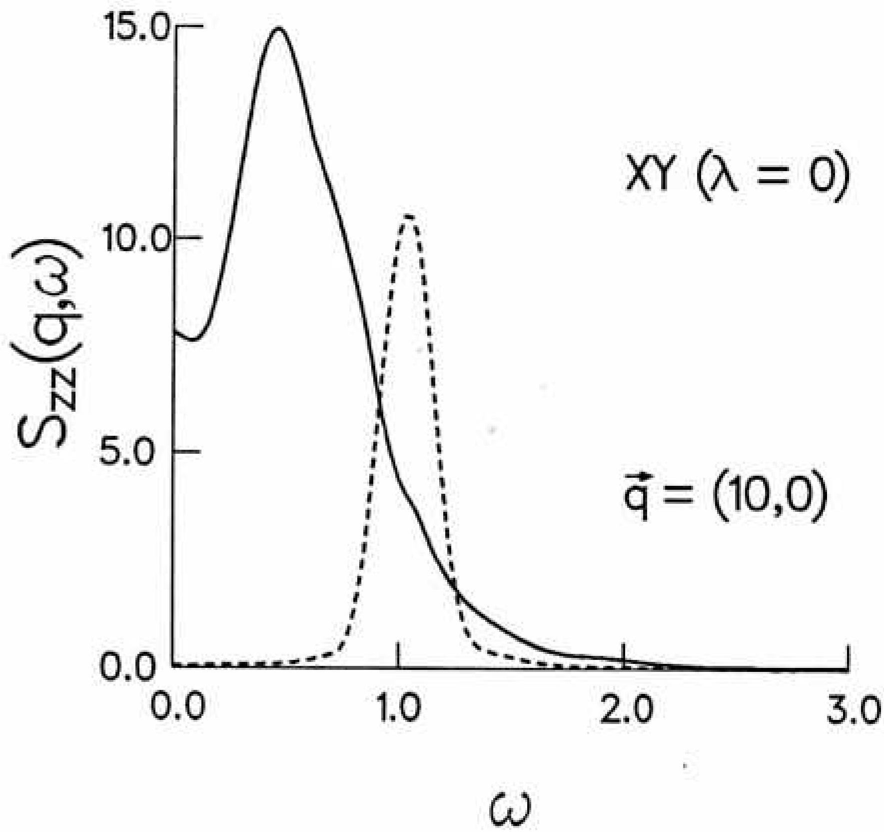
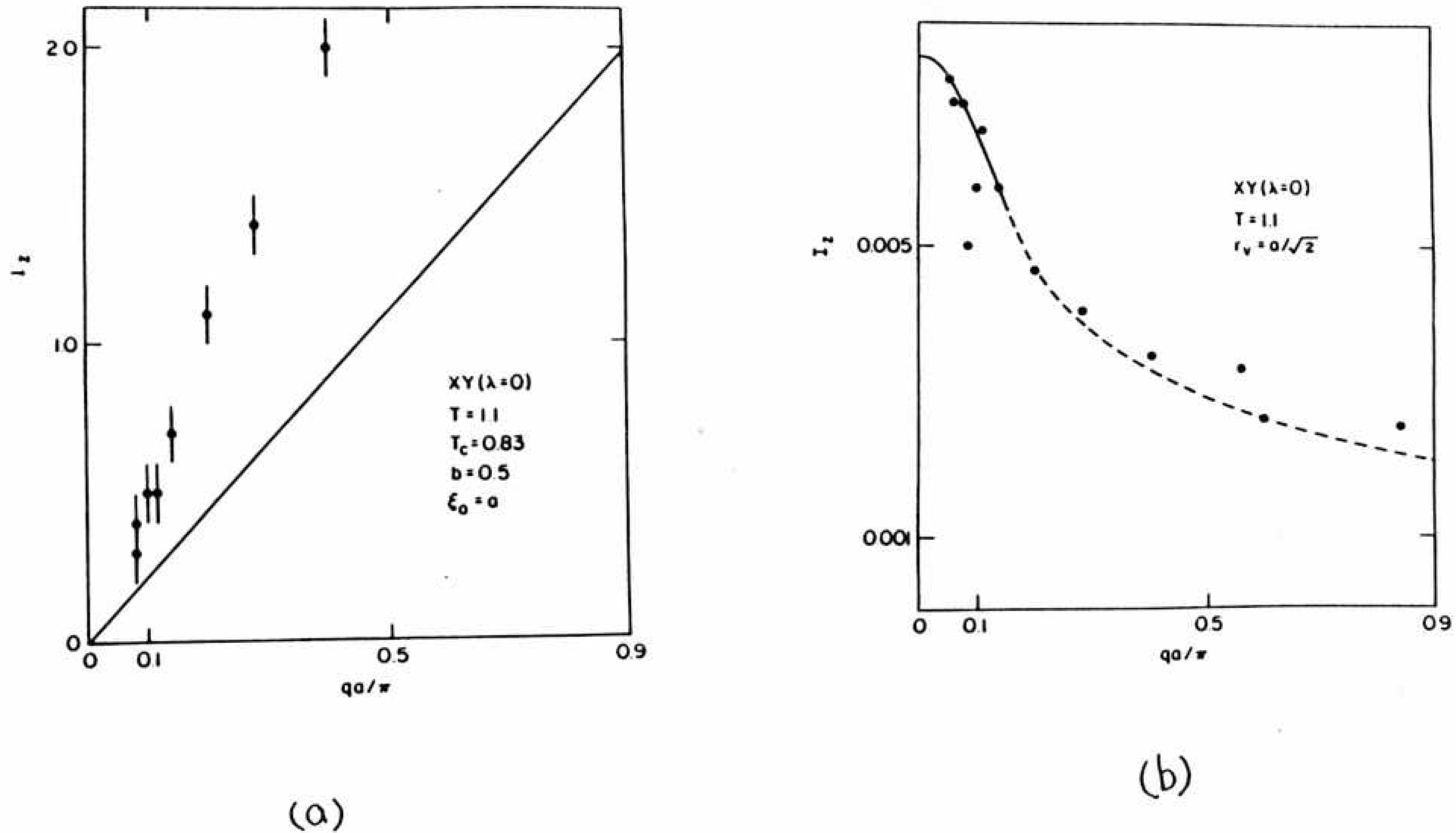


FIG 1



I_z
20
10
0
0
0.1
0.5
0.9
qa/π
XY(λ=0)
T=1.1
T_c=0.83
b=0.5
ξ_0=a
(a)
I_z
0.005
0.001
0
0.1
0.5
0.9
qa/π
XY(λ=0)
T=1.1
r_v=a/√2
(b)

FIG. 2

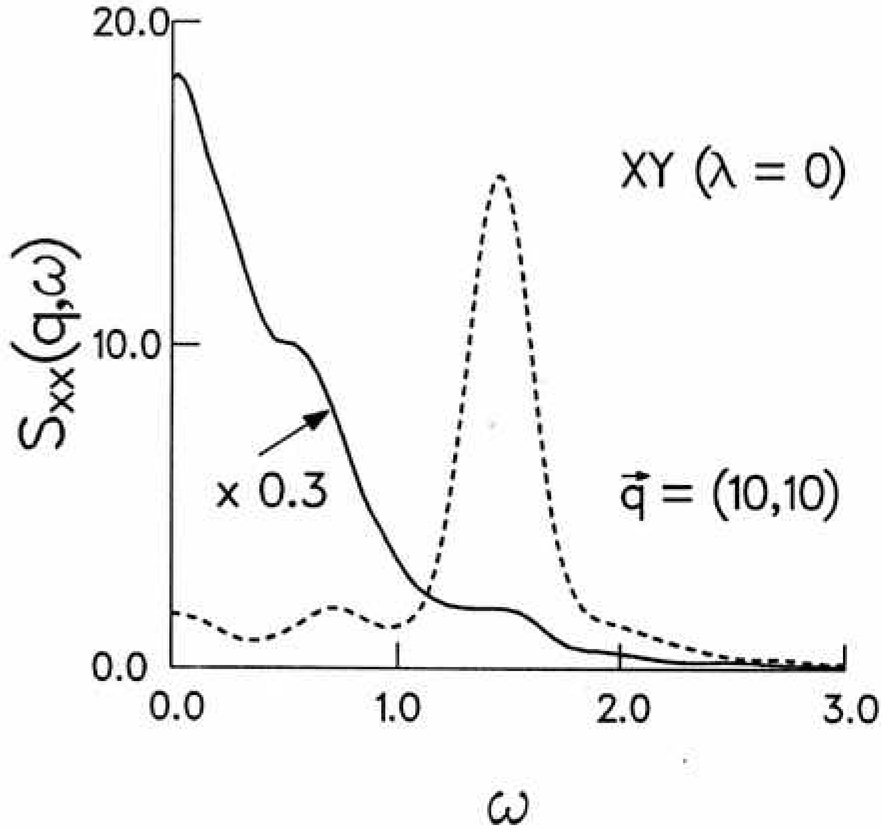
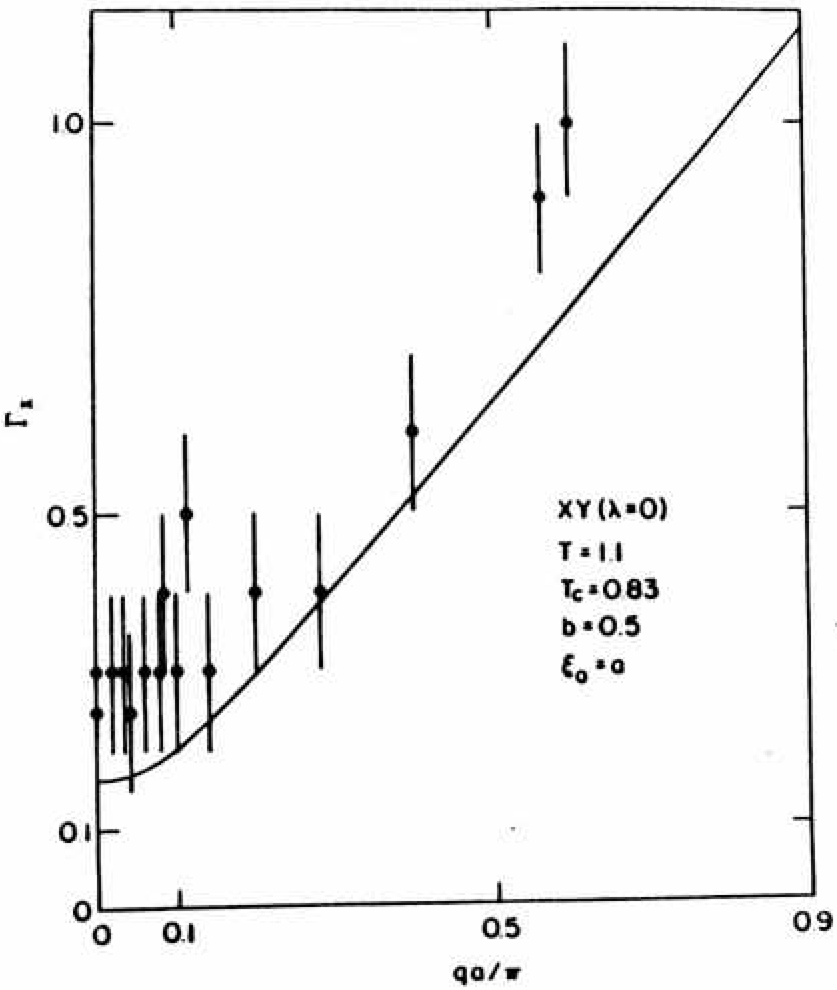
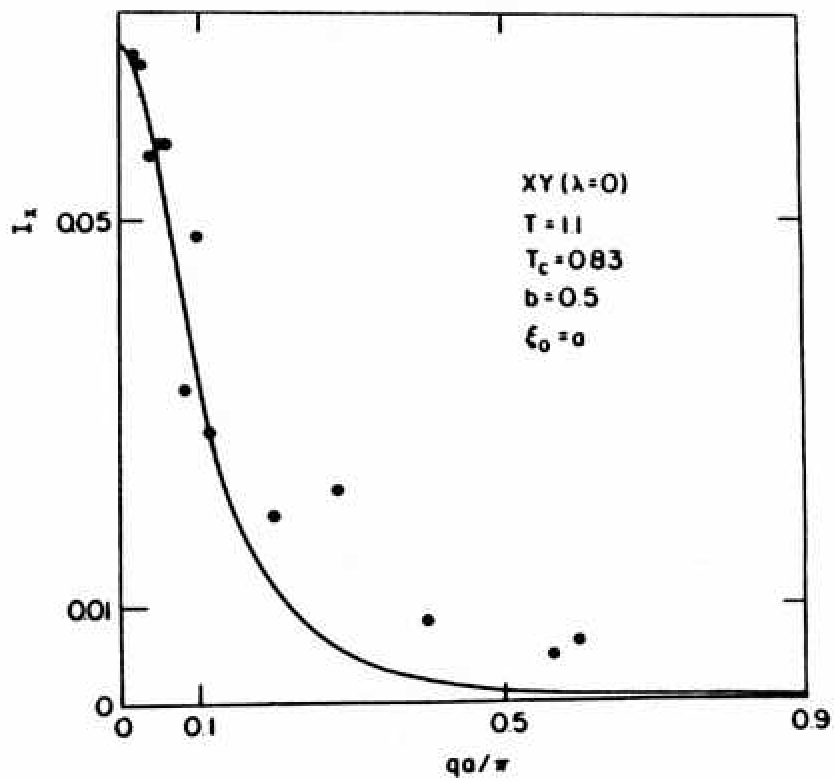

20.0
10.0
0.0
0.0
1.0
2.0
3.0
$S_{xx}(q,\omega)$
ω
XY ($\lambda = 0$)
$\vec{q} = (10,10)$
x 0.3

FIG 3



1.0
0.5
0.1
0
0
0.1
0.5
0.9
qa/π
Γx
XY (λ=0)
T = 1.1
Tc = 0.83
b = 0.5
ξ0 = a

(a)



0.05
0.01
0
0
0.1
0.5
0.9
qa/π
Ix
XY (λ=0)
T = 1.1
Tc = 0.83
b = 0.5
ξ0 = a

(b)

FIG. 4