A theory of spin correlations and neutron scattering from paramagnetic materials based on the Ising - Heisenberg model in one, two and three space dimensions

1996 J. Phys.: Condens. Matter 8 L307

(http://iopscience.iop.org/0953-8984/8/21/001)

J. Phys.: Condens. Matter **8** (1996) L307–L310. Printed in the UK

LETTER TO THE EDITOR

A theory of spin correlations and neutron scattering from paramagnetic materials based on the Ising–Heisenberg model in one, two and three space dimensions

Stephen W Lovesey
ISIS Facility, Rutherford Appleton Laboratory, Oxon OX11 0QX, UK

Received 7 February 1996

Abstract. Responding to the potential of a new neutron-beam method for studying the time-dependent properties of magnetic fluctuations, well suited for quasi-two-dimensional magnets, we predict the form of the signal for scattering by paramagnetic materials. The model used is expected to describe Rb_2CoF_4 and related quasi-two-dimensional magnets, with an Ising-like character. Calculations are made using the coupled-mode theory of spin fluctuations. Applied to an isotropic Heisenberg model, results from the theory are not the same as those stemming from Fick's Law for spin diffusion . For Rb_2CoF_4 we predict out-of-plane fluctuations to be the same as for the Heisenberg model, and in-plane fluctuations to be different and an exponential function of time.

There is currently great interest in the properties of various models of magnetic systems with the common feature of magnetic moments located at the sites of a planar lattice. Several decades ago interest in planar, or equivalently two-dimensional, models underwent a sharp revision following Onsager's demonstration that an Ising model on a square planar lattice shows a spontaneous, long-range ordering of the moments at a sufficiently low temperature, proportional to the strength of the exchange interaction between adjacent moments [1]. Later it was proven that a Heisenberg model, with a fully isotropic exchange interaction, on a planar lattice and at a non-zero temperature does not support long-range order. The same is true of the plane-rotator model, in which spin fluctuations are constrained to lie in a plane, although for this model the static susceptibility, specific heat etc, diverge at a non-zero temperature through the spontaneous generation of vortex (topological) excitations. Interest in planar models has been propelled to its current intensity largely by the discovery of ceramic superconductors, with relatively high critical temperatures, since most of the materials also display magnetic properties which can be interpreted in terms of the properties of copper ions arranged on sheets. In summary, today, the static and time-dependent features of both classical [2] and quantum [3] versions of magnetic models on planar lattices are a main theme in statistical mechanics.

A useful addition to the range of accurate experimental methods for the study of quasi-two-dimensional magnets has recently been successfully demonstrated on a sample of Rb_2CoF_4 diluted with non-magnetic ions [4]. The method appears to give direct access to spin autocorrelation functions central in the development of our understanding of the time-dependent properties of planar magnets. The goal of the work reported in this letter is to predict the findings of an experiment of the type in question on an undiluted sample of Rb_2CoF_4 or a similar compound [5]. In the course of achieving this, we also provide results for magnetic models with moments arranged on a chain and on cubic lattices.

0953-8984/96/210307+04$19.50

Regrettably, perhaps, most quasi-two-dimensional magnetic materials are not adequately described by an Ising model or a Heisenberg model, and more complicated exchange interactions are required. In the case of Rb_2CoF_4 a suitable model is a weighted sum of Ising and Heisenberg interactions [6] and it is this model we analyse in the limit of a high temperature. Our theoretical work uses the coupled-mode theory for time-dependent properties, which furnishes a precise description of magnetic fluctuations with a very long wavelength. Results obtained with this theory are different from those stemming from Fick's Law for spin diffusion in paramagnets [7].

The magnetic model has spin operators $\{S(\boldsymbol{R})\}$ attached to the sites $\{\boldsymbol{R}\}$ of a lattice with spatial dimension $d = 1$, 2 or 3. Spins on adjacent sites interact via a sum of an Ising exchange, $S^z(\boldsymbol{R})S^z(\boldsymbol{R}')$, and a Heisenberg exchange, $S(\boldsymbol{R}) \cdot S(\boldsymbol{R}')$, and the corresponding exchange parameters are $(I - J)$ and J. Our definitions of the parameters in the model make it the same as the one used to analyse excitations in Rb_2CoF_4 [6], for which $(J/I) = 0.55$. In the limit of a high temperature, much in excess of maximum $(|I|,\ |J|)$, static and dynamic properties of the model do not depend on the signs of I and J.

In the limit of a high temperature, the cross-section for the inelastic scattering of neutrons, which suffer an energy change ω and wavevector change $\boldsymbol{Q}$, is proportional to the spin relaxation functions

$$\sum_{\alpha,\beta} \left(\delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right) R^{\alpha\beta}(\boldsymbol{Q},\ \omega) \tag{1}$$

where $\hat{\boldsymbol{Q}} = \boldsymbol{Q}/Q$. In (1), $R^{\alpha\beta}(\boldsymbol{Q},\ \omega)$ is the Kubo relaxation function formed with spatial Fourier components of the spin operators $S^\alpha(\boldsymbol{R})$ and $S^\beta(\boldsymbol{R}')$. For a uniaxial magnet, $R^{\alpha\beta}(\boldsymbol{Q},\ \omega) = 0$ if $\alpha \neq \beta$, and $R^{xx}(\boldsymbol{Q},\ \omega) = R^{yy}(\boldsymbol{Q},\ \omega) = R_0(\boldsymbol{Q},\ \omega)$, say, and let $R^{zz}(\boldsymbol{Q},\ \omega) = R(\boldsymbol{Q},\ \omega)$. The cross-section is thus proportional to,

$$\left(1 - \hat{Q}_z^2\right) R(\boldsymbol{Q},\ \omega) + \left(1 + \hat{Q}_z^2\right) R_0(\boldsymbol{Q},\ \omega). \tag{2}$$

An integration of the cross-section with respect to $\boldsymbol{Q}$ gives direct access to power spectra of the time-dependent spin autocorrelation functions,

$$G^{\alpha\alpha}(\boldsymbol{R},\ t) = \left\langle S^\alpha\left(\boldsymbol{R}_0,\ 0\right) S^\alpha\left(\boldsymbol{R} + \boldsymbol{R}_0,\ t\right)\right\rangle \tag{3}$$

where the angular brackets denote a thermal average of the enclosed operators. In the experiment described in [4] the aim is to have a signal proportional to the power spectrum of $G^{\alpha\alpha}(\boldsymbol{R} = 0,\ t)$, in which the conjugate variable is the change in energy, ω.

The spin correlation functions, mentioned in the foregoing paragraph, are obtained theoretically from a study of the equations of motion, for relaxation functions $F(\boldsymbol{q},\ t)$ and $F_0(\boldsymbol{q},\ t)$, derived from the Hamiltonian. The equations are part of a hierarchy of equations which is unbounded, and so not all properties can be calculated without approximations. We are confident in our method, often called the coupled-mode theory, for a precise description of properties at long wavelengths. Central to the implementation of the chosen method are memory or self-energy functions denoted by $K(\boldsymbol{q},\ t)$ and $K_0(\boldsymbol{q},\ t)$ for the out-of-plane and in-plane fluctuations, respectively. The Laplace transforms of $F(\boldsymbol{q},\ t)$ and $K(\boldsymbol{q},\ t)$ satisfy

$$\tilde{F}(\boldsymbol{q},\ s) = \{s + \tilde{K}(\boldsymbol{q},\ s)\}^{-1} \tag{4}$$

and the quantity in the cross-section

$$R(\boldsymbol{q},\ \omega) = (1/\pi)\mathrm{Re}\ \tilde{F}(\boldsymbol{q},\ \mathrm{i}\omega). \tag{5}$$

All our findings are derived from the results

$$K(\boldsymbol{q},\ t) \propto q^2 \int_q p^{d+1}\,\mathrm{d}p F_0^2(\boldsymbol{p},\ t) \tag{6}$$

and

$$K_0(\boldsymbol{q},\, t) \propto \zeta^4 \int_q p^{d-1}\, \mathrm{d}p F_0(\boldsymbol{p},\, t) F(\boldsymbol{p},\, t) \tag{7}$$

which are obtained by application of the standard coupled-mode theory to our Ising–Heisenberg model, evaluated in the limit of a high temperature and a very small wavevector q. The wavevector ζ is a measure of the difference in the Ising and Heisenberg exchange parameters, specifically $(I - J)^2 \propto \zeta^4$.

$F(\boldsymbol{q},\, t)$ and $K(\boldsymbol{q},\, t)$ and the corresponding quantities for in-plane spin fluctuations, are found to be homogeneous functions of the form (λ arbitrary)

$$F(\boldsymbol{q},\, t) = F\big(q\lambda^a,\, t\lambda^b,\, \zeta\lambda^c\big) \tag{8}$$

and

$$K(\boldsymbol{q},\, t) = \lambda K\big(q\lambda^a,\, t\lambda^b,\, \zeta\lambda^c\big) \tag{9}$$

in which $a = c = -1/(2\theta)$ with $\theta = (4 + d)/2$, and $b = 1/2$ follows directly from (4). The next steps are to set $\zeta\lambda^a = 1$, and analyse the equations for $K(q,\, t)$ and $K_0(q,\, t)$ in the limit $(q/\zeta) \to 0$.

The z-component of the spin density is a constant of the motion and, unlike the Zeeman energy induced by a magnetic field, the Ising exchange energy remains unchanged on reversal of the direction of the spin variables. In the limit of $q \ll \zeta$, $K(q,\, t)$ will evolve with time in a manner independent of ζ. This condition on $K(q,\, t)$ leads to the result, valid for long times,

$$K(q,\, t) \propto q^2 (1/t)^{(d+2)/\theta}. \tag{10}$$

With $1 \leqslant d \leqslant 3$ the exponent of $(1/t)$ in (10) is larger than one and less than two, and it follows that $F(q,\, t)$ and $K(q,\, t)$ have a common asymptotic dependence on $(1/t)$. $F(q,\, t)$ is a function of the single variable tq^θ and this prompts us to write

$$F(q,\, t) = f\big(tq^\theta\big). \tag{11}$$

For the spin correlation function in real space

$$G(\boldsymbol{R} = 0,\, t) = \big\langle S^z\big(\boldsymbol{R}_0,\, 0\big) S^z\big(\boldsymbol{R}_0,\, t\big)\big\rangle \propto \int p^{d-1}\, \mathrm{d}p\; F(p,\, t) \propto (1/t)^{d/\theta}. \tag{12}$$

Since $(d/\theta) < 1$ the power spectrum of $G(0,\, t)$ is proportional to $(1/\omega)^{1-d/\theta}$ for $\omega \to 0$.

A corresponding analysis of $K_0(q,\, t)$ leads us to conclude it is independent of q for $q \ll \zeta$. This finding implies that there is no correlation between the in-plant spin components on different lattice sites, and $G_0(\boldsymbol{R},\, t) = 0$ for $\boldsymbol{R} \neq 0$. In consequence

$$F_0(q,\, t) = f_0\big(t\zeta^\theta\big) \propto G_0(0,\, t). \tag{13}$$

The function $f_0(t)$ is found to be an exponential function of t. This last result is a self-consistent solution of (7) and the analogue of (4) for in-plane fluctuations, taken together with the result in (12). Writing

$$f_0(t) \propto \exp\big(-\Gamma_0 t\big) \tag{14}$$

the decay rate Γ_0 is independent of ζ, and the power spectrum of $G_0(0,\, t)$ is a Lorentzian function of ω with a width Γ_0. By way of comment on our finding for $F_0(q,\, t)$ we report the corresponding result for an isotropic Heisenberg magnet in a magnet field which produces a Zeeman energy

$$-H \sum_{\boldsymbol{R}} S^z(\boldsymbol{R}).$$

In the limit $q \to 0$ we find $F_0(q, t)$ is independent of q and

$$F_0(q, t) = J_1(2Ht)/(Ht)$$

where $J_1(x)$ is a Bessel function of order one. From (5)

$$R_0(q, \omega) = \left(4H^2 - \omega^2\right)^{1/2} / \left(2\pi H^2\right) \qquad \omega^2 < 4H^2$$
$$= 0 \qquad \text{otherwise.}$$

Hence the properties of $F_0(q, t)$ for the two models, namely a Heisenberg magnet plus a magnetic field or an Ising exchange, are quite similar.

With regard to the neutron scattering cross-section our results lead to the following predictions. If (2) is integrated with respect to $\boldsymbol{q}$ the signal observed is a weighted sum of the power spectra of $G(0, t)$ and $G_0(0, t)$, e.g. the out-of-plane power spectrum

$$\int_{-\infty}^{\infty} \mathrm{d}t\, G(0, t) \exp(-\mathrm{i}\omega t).$$

For small ω, the out-of-plane and in-plane spectra are proportional to $(1/\omega)^{1-d/\theta}$ and a Lorentzian function of ω, respectively.

It might be useful to look at the corresponding predictions based on Fick's Law for spin diffusion. For a pure Heisenberg model $(I = J)$, Fick's Law leads to the result

$$F(q, t) \propto \exp\left(-Dq^2 t\right)$$

where D is a diffusion constant, and hence

$$G(0, t) \propto (1/t)^{d/2}.$$

On the other hand, use of coupled-mode theory gives[7]

$$G(0, t) \propto (1/t)^{d/\theta}$$

and this is the same result we have obtained for out-of-plane fluctuations in the Ising–Heisenberg model.

The result for $G(0, t)$ comes from integrating $F(\boldsymbol{q}, t)$ with respect to $\boldsymbol{q}$. For $d \geqslant 2$ one might consider integration with respect to one component of $\boldsymbol{q}$. Use of Fick's Law for $d = 2$ gives

$$\int \mathrm{d}q_x\, F(\boldsymbol{q}, t) \propto (1/t)^{1/2} \exp\left(-Dq_y^2 t\right)$$

while coupled-mode theory for the same quantity predicts a long time dependence of the form $(1/t)^{1/\theta}$. These results show that the observed power spectrum depends on the number of components of $\boldsymbol{q}$ integrated in the execution of the experiment.

The author has benefitted from discussions with M A Adams, H Ikeda, and P Verrucchi.

References

[1] For a review of this and related work see, for example, Mattis D C 1985 *The Theory of Magnetism II* (Berlin: Springer)
[2] Curély J 1995 *Europhys. Lett.* **32** 529
[3] Auerbach A 1994 *Interacting Electrons and Quantum Magnetism* (New York: Springer)
[4] Ikeda H, Itoh S and Adams M A 1995 *Phys. Rev. Lett.* **75** 4440
[5] Collins M F 1989 *Magnetic Critical Scattering* (New York: Oxford University Press)
[6] Ikeda H and Hutchings M T 1978 *J. Phys. C: Solid State Phys.* **11** L529
[7] Lovesey S W, Engdahl E, Cuccoli A, Tognetti V and Balcar E 1994 *J. Phys.: Condens. Matter* **6** L521