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LETTER TO THE EDITOR

Critical spin dynamics of EuO: comparison of theory and experiment

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Abstract. The standard coupled-mode theory of spin dynamics is evaluated for an isotropic, Heisenberg model of EuO at the critical temperature. The predicted long-wavelength limit of the spin-relaxation function is approximately a gaussian function of time in the time interval probed in recent neutron spin-echo measurements. This functional form is distinctly different from the observed, almost exponential, decay.

Our current understanding of critical spin dynamics of Heisenberg magnets is based essentially on two theoretical approaches. Renormalisation group calculations, using continuum versions of the Heisenberg magnet, provide firmly based insights into the nature of spin-correlation functions in the critical region. In particular, the calculations vindicate the dynamic scaling hypothesis and give values for critical exponents; for reviews see, for example, Brézin and Parisi (1978), Hohenberg and Halperin (1977) and Kawasaki and Gunton (1976). Another approach, devised before the development of the renormalisation group, relies on approximate self-consistent equations derived from the spin-operator equation of motion. For largely historic reasons, these self-consistent equations are usually called the coupled-mode theory, and different derivations are given by Hubbard (1971), Kawasaki (1975) and Wegner (1968). The coupled-mode theory is consistent with the dynamic scaling hypothesis, and predicted critical exponents agree with results from renormalisation group calculations.

The coupled-mode theory affords an explicit prediction of the spin-autocorrelation function whose temporal Fourier transform is observed in inelastic magnetic neutron scattering. Comparisons of theory and experimental results for ferromagnets, both at and above the critical temperature, provide strong support in favour of the coupled-mode theory. Similar support is provided by computer simulations (Takahashi 1983).

For all the reasons outlined in the preceding paragraphs, the coupled-mode theory of Heisenberg ferromagnets is generally regarded as very reliable. This view is challenged by Mezei (1985) on the basis of precise measurements of the spin-correlation function of the insulating ferromagnet EuO at the critical temperature. The measured correlation function is consistent with a simple exponential time decay, proposed in the conventional Van Hove theory of critical slowing down, and thus apparently at odds with results deduced from the Wegner (1968) and Hubbard (1971) coupled-mode calculations. We report a detailed analysis of the coupled-mode theory for EuO, using an isotropic Heisenberg model, which exposes differences between theory and experimental results that are even more pronounced than implied by Mezei's initial analysis. In the time interval probed in the experiments, the coupled-mode spin-correlation function is a gaussian function of time, to a good approximation, whereas the data are consistent with an exponential decay, as mentioned already.

Let us begin with a brief survey of the coupled-mode theory of spin correlations in the Heisenberg magnet described by the Hamiltonian

$$\mathcal{H} = -\sum_{n,m} J(\boldsymbol{n} - \boldsymbol{m})\boldsymbol{S}_n \cdot \boldsymbol{S}_m. \tag{1}$$

Here, $J(\boldsymbol{m})$ is the exchange coupling between spins S located at sites defined by lattice vectors $\boldsymbol{m}$, and $J(0) = 0$; values of J for EuO have been obtained by Passell *et al* (1976). The spectrum of spontaneous spin fluctuations observed in inelastic magnetic neutron scattering is

$$\begin{aligned} S(k, \omega) &= \int_{-\infty}^{\infty} \frac{\mathrm{d}t}{2\pi} \exp(-\mathrm{i}\omega t)\langle S^\alpha(-\boldsymbol{k})S^\alpha(\boldsymbol{k}, t)\rangle \\ &= \chi(k)\omega(1 + n(\omega)) \int_{-\infty}^{\infty} \frac{\mathrm{d}t}{2\pi} \exp(-\mathrm{i}\omega t)F(k, t). \end{aligned} \tag{2}$$

For an isotropic spin system, the correlation function is independent of the Cartesian label α. The second equality in (2) expresses $S(k, \omega)$ in terms of the normalised spin-relaxation function $F(k, t) = F(k, -t)$, and the wave-vector-dependent susceptibility $\chi(k)$. The detailed balance factor is written in terms of $n(\omega) = [\exp(\omega/T) - 1]^{-1}$ where T is the temperature, using units with $\hbar = k_\mathrm{B} = 1$.

With the coupled-mode theory $F(k, t)$ is obtained from the equations

$$\partial_t F(k, t) = -\int_0^t \mathrm{d}t'\, K(k, t - t')F(k, t') \tag{3}$$

and

$$K(k, t) = \frac{8T}{\chi(k)\lambda} \sum_{q} (\mathcal{J}(\boldsymbol{q}) - \mathcal{J}(\boldsymbol{k} - \boldsymbol{q}))\, \frac{F(k - q, t)F(q, t)}{\mu - \lambda\mathcal{J}(\boldsymbol{q})} \tag{4}$$

where

$$\mathcal{J}(k) = \sum \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{m})J(\boldsymbol{m}).$$

The parameters μ and λ appear in the susceptibility

$$\chi(k) = N/(\mu - \lambda\mathcal{J}(\boldsymbol{k})) \tag{5}$$

which is derived from the coupled-mode theory by requiring it to satisfy the f-sum rule. At the ferromagnetic critical temperature $\mu = \lambda\mathcal{J}(0)$. We take $\lambda = 2$ since then (5) is exact in the limit $T \to \infty$, and the observed critical temperature is consistent with the estimate

$$\lambda^2\mathcal{J}(0)S(S + 1)/T_\mathrm{c} = (6/N) \sum_{q} (1 - \mathcal{J}(\boldsymbol{q})/\mathcal{J}(0))^{-1} \tag{6}$$

obtained from (5) and the identity $\boldsymbol{S} \cdot \boldsymbol{S} = S(S + 1)$.

At the critical temperature, and in the limit $k \to 0$, we find ($\tau \geq 0$)

$$F(k, \tau/\theta) = \frac{Q(\tau)}{2\pi} = \int_{-\infty}^{\infty} \frac{\mathrm{d}\varepsilon}{2\pi} \frac{\exp(\mathrm{i}\varepsilon\tau)}{\mathrm{i}\varepsilon + g(\varepsilon)} \tag{7}$$

and

$$\int_0^{\infty} \mathrm{d}t \exp(-\mathrm{i}\omega t) K(k, t) = \theta g(\omega/\theta) \tag{8}$$

where

$$g(\varepsilon) = \frac{2}{3} \int_0^{\infty} \mathrm{d}\tau \exp(-\mathrm{i}\varepsilon\tau)(Q(\tau))^2. \tag{9}$$

In these expressions the frequency $\theta = \xi k^{5/2}$ and ξ is related to the exchange couplings by the $k \to 0$ limit of

$$\xi^2 = v_0(\mathcal{J}(0) - \mathcal{J}(k))T_c/2k^2\pi^4 \qquad k \to 0$$

in which v_0 is the unit-cell volume. For EuO we obtain

$$\xi^2 = a^5(J_1 + J_2)T_c/8\pi^4 = 1.72\,\mathrm{meV}^2\,\text{Å}^5$$

in which J_1 and J_2 are first- and second-neighbour exchange couplings, and $a = 5.14$ Å is the cube edge in the FCC lattice.

Equations (7) and (9) are sufficient to solve for the functions $Q(\tau)$ and $g(\varepsilon)$. In order to solve them numerically, we cut off the integrals at $\varepsilon = E$ and $\tau = T$, where E and T are large, so (7) can be written

$$\tfrac{1}{2}Q(\tau) = \int_0^{E} \mathrm{d}\varepsilon \, \mathrm{Re}\left(\frac{\mathrm{e}^{\mathrm{i}\varepsilon\tau}}{\mathrm{i}\varepsilon + g(\varepsilon)}\right) + \int_{E\tau}^{\infty} \mathrm{d}x \frac{\sin(x)}{x}. \tag{10}$$

We compute the integrals by the trapezoidal rule with N strips, so $\delta\varepsilon = E/N$ and $\delta\tau = T/N$, and use a standard approximation for the sine integral in (10). We start the iteration with $g(\varepsilon) = 1$ in (10), obtain $Q(\tau)$, and get a new $g(\varepsilon)$ from (9). To check the procedure we calculated $g(\varepsilon)$ to the second iteration analytically, and this was within 10^{-4} of the numerical result with $N = 100$, $E = 50$ and $T = 2$. After seven iterations with these values, convergence was achieved, and $Q(\tau)$ is illustrated in figure 1 (curve A). Increasing N, E and T does not affect this curve, and neither does the initial choice for $g(\varepsilon)$; although there is an oscillation of wavelength $2\pi/E$ discernible in $Q(\tau)$ due to the cut-off in $g(\varepsilon)$, which is expected when a discontinuity is introduced in a Fourier transform. The value of $g(0)$ is 4.510, and for $\omega \ll \theta < T_c = 5.95$ meV the spectrum $S(k, \omega)$ approximates to a Lorentz function of ω with a half-width $= \theta g(0)$.

To supplement the numerical solution, we can derive an asymptotic solution to the coupled equations (7) and (9) as follows. Let $s = 4\pi/3^{1/2}\mathrm{i}\varepsilon$ and $\gamma = (\tau\omega_0/\theta)^2/2$, where $\omega_0 = \theta 2\pi(\frac{2}{3})^{1/2}$ is the exact value of the frequency that occurs in the f-sum rule

$$\int_{-\infty}^{\infty} \mathrm{d}\omega\, \omega S(k, \omega) = \chi(k)\omega_0^2 = -\chi(k)\partial_t^2 F(k, t)|_0. \tag{11}$$

Suppose we can write

$$(\mathrm{i}\varepsilon + g(\varepsilon))^{-1} = (\theta/2^{1/2}\omega_0) \sum_{l=0}^{\infty} b_l s^{2l+1}. \tag{12}$$

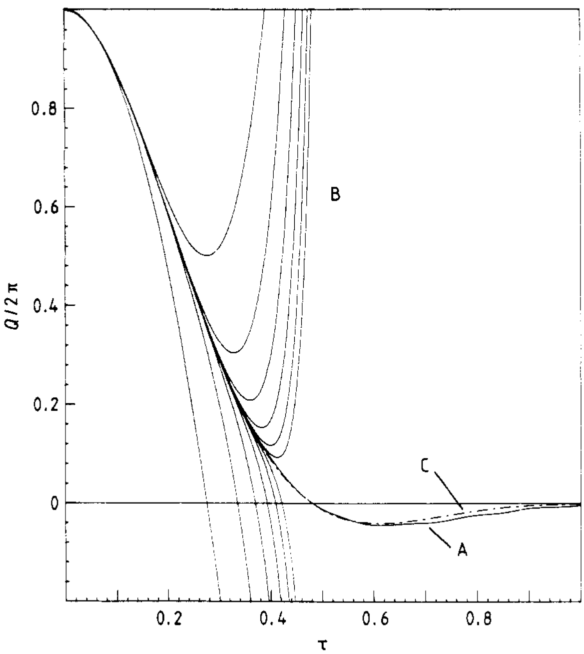

Figure 1. Values of $Q(\tau)/2\pi$ computed by various methods are shown for $0 < \tau < 1.0$. Curve A is the complete numerical solution of equations (7) and (9) obtained with the technique described in the text. Curves B are a family of results derived from (13) with an increasing number of terms. Curve C is obtained from the asymptotic series (16).

Then from (7) and (9)

$$Q(\tau) = 2\pi \sum_{l=0}^{\infty} b_l (4\gamma)^l/(2l)! \tag{13}$$

$$g(\varepsilon) = (\omega_0/2^{1/2}\theta) \sum_{l=0}^{\infty} c_l s^{2l+1} \tag{14}$$

where the coefficients b_l and c_l satisfy

$$b_0 = 1 \qquad c_l = \sum_{m=0}^{l} \binom{2l}{2m} b_m b_{l-m} \qquad 2b_{l+1} = -\sum_{m=0}^{l} c_m b_{l-m}. \tag{15}$$

The first few coefficients b_l and c_l are listed in table 1. In figure 1 (curves B) we show the first 12 partial sums of the series (13) for $Q(\tau)$, and it is clear that the series asymptotically approaches the numerical solution.

Table 1. Coefficients in the expansion of $Q(\tau)$ and $g(\varepsilon)$, and those for a gaussian $g(\varepsilon)$.

l	b_l	c_l	c'_l
0	1	1	1
1	−0.5	−1	−1
2	0.75	3	3
3	−2.125	−15.5	−15
4	9.9375	118.75	105
5	−70.40625	−1257.375	−945

For small τ, $Q(\tau)$ looks gaussian, and if it were, the real part of $g(\varepsilon)$ would be gaussian, and the imaginary part proportional to Dawson's integral. The asymptotic series for $g(\varepsilon)$ would have the coefficients c_l replaced by c_l' (see table 1), which are Hermite polynomials of argument zero, $c_l' = H_{2l}(0)/2^l$. The two series c_l and c_l' are the same up to $l = 2$, with progressively larger deviations, so we can write

$$Q(\tau) = 2\pi \exp(-\gamma)[1 - (\gamma^3/45) - (\gamma^4/1260) - (23\gamma^5/18900)]. \qquad (16)$$

The sum to the γ^5-term is closest to the numerical solution, and is drawn as a chain curve in figure 1 (curve C); this serves as a convenient numerical approximation to $Q(\tau)$.

Mezei (1985) reports measured values of $F(k, t)$ for $T = T_c$, $k = 0.024$ Å^{-1} and $0 \leq t \leq 2$ ns that are tolerably well represented by a single exponential with a decay time of 0.83 ns. The measured t-dependence of $F(k, t)$ is thus distinctly different from that predicted by the coupled-mode theory. Using the value $(1/\theta) = 5.5$ ns, appropriate for EuO and $k = 0.024$ Å^{-1}, we find $F(k, t = 1 \text{ ns}) = 0.65$ whereas Mezei's result is about half this value.

The discrepancy between theory and experiment is pronounced at short times. The exact short-time behaviour of the relaxation function is

$$F(k, t) = 1 - (t\omega_0)^2/2 + \ldots$$

which is in stark contrast with the observed, essentially exponential, decay. For large times, such that $\theta t \gg 1$, theory predicts an exponential tail to the relaxation function of the form $\exp(-\theta g(0)t)$. In seeking reasons for the discrepancy, other than inadequacy of the coupled-mode theory or erroneous data interpretation, we are led to question the possible importance of additional terms in the Hamiltonian. However, the most likely candidate, a dipolar interaction, has been shown by Mezei (1984) to be unimportant at $k = 0.024$ Å^{-1} and $T = T_c$.

F Mezei kindly made his results available prior to publication, and commented on our findings. We have benefited from conversations with U Balucani and G Shirane in the course of our work.

References

Brézin E and Parisi G 1978 *J. Stat. Phys.* **19** 269
Hohenberg P C and Halperin B I 1977 *Rev. Mod. Phys.* **49** 435
Hubbard J 1971 *J. Phys. C: Solid State Phys.* **4** 53
Kawasaki K 1975 *Phase Transitions and Critical Phenomena* vol 5a, ed. C Domb and M S Green (London: Academic)
Kawasaki K and Gunton J D 1976 *Progress in Liquid Physics* ed. C Croxton (Colchester: Wiley)
Mezei F 1984 *J. Magn. Magn. Mater.* **45** 67
—— 1985 *Critical Dynamics in EuO at the Ferromagnetic Curie Point* to be published
Passell L, Dietrich O W and Als-Nielsen J 1976 *Phys. Rev.* B **14** 4897
Takahashi M 1983 *J. Phys. Soc. Japan* **52** 3592
Wegner F 1968 *Z. Phys.* **216** 433