Critical Scattering and Dynamical Scaling in an Heisenberg Ferromagnet
Neutron Spin Echo versus Renormalization Group Theory

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High resolution Neutron Spin Echo (NSE) spectroscopy was used to investigate the dynamics of an 3D Heisenberg ferromagnet in the exchange-controlled regime over a broad range of temperatures and momentum transfer. These results allow for the first time an extensive comparison between the experimental dynamical critical behavior and the predictions of the Renormalization Group (RG) theory. The agreement is exhaustive and surprising as the RG theory accounts not only for the critical relaxation but also for the shape crossover towards an exponential diffusive relaxation when moving from the critical to the hydrodynamic regime above $T_C$.

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Critical phenomena are ubiquitous in condensed matter physics and their theories are now used to understand the behavior of many complex systems, like sandpiles or tropical rains. Phase transitions are dynamical processes in essence, specially the continuous phase transitions. These are characterized by diverging fluctuations at the critical temperature $T_C$, with a critical slowing-down and a scale-free behavior: power law spatial and temporal evolutions of the order-parameter fluctuations. In the critical region, the dynamics is governed by a single dimensionless variable, according to the dynamical scaling hypothesis. Ferromagnetism serves as a paradigm for continuous phase transitions and as such a lot of theoretical and experimental effort has been devoted to understand and predict in details its static and dynamic critical behavior.

Critical dynamics is a much more complex problem than the static critical phenomena as it depends not only on the static universality class but also on conservation laws and on the equations of motion of the system. For example, the 3D Heisenberg ferromagnet displays two different behaviors, depending on the relevant interactions, exchange or dipolar. The balance between exchange or dipolar interactions is quantified by the dipolar wave-vector $Q_D (Q_D^2 = \mu_0 k_B T \mu_B^2 J_2^2)$, $J_2$ being the second moment of the exchange interactions. Exchange controls the dynamical behavior for $Q > Q_D$ and the order-parameter (i.e. the magnetization) is conserved (model $J$ of Reference (7)). The critical dynamics at $T_C$ is governed by a dynamical exponent $z=\frac{D+2-\eta}{2}$, where $D$ is the space dimension and $\eta$ the Fisher exponent. For 3-D Heisenberg systems, $z \approx 5/2$ (figure 1). Dipolar interactions, on the other hand, control the dynamics at $Q < Q_D$. These interactions do not conserve the order parameter and at $T_C$ the dynamical exponent crosses over from $z=5/2$ (exchange) to $z=2$ (dipolar) at $Q \approx Q_D/10$.

![FIG. 1: Critical slowing down of the characteristic relaxation time $\tau_c$ of the magnetic fluctuations in CdCr$_2$S$_4$. The solid line is a power law fit, $\tau_c(Q,T_C) = \tau_0 Q^{-z}$ with $z = 2.47 \pm 0.02$ and $\tau_0 = 0.02813 \text{ ns/nm}^{-z}$.](image-url)

In the critical region above $T_C$ and in the sole presence of exchange interactions, the relevant scaling variable is $x = \kappa/Q$, with $\kappa$ the inverse magnetic correlation length. In this region the magnetic fluctuations decay faster than an exponential with a shape cross-over to an exponential relaxation when going from the critical to the hydrodynamic regime for $x \gg 1$. This simple scaling hypothesis...
breaks down in the presence of dipolar interactions. At $Q < Q_D$, the relaxation time depends on two reduced variables: $x = \kappa/Q$ and $y = Q/Q_D$ [8] whereas the relaxation recovers a simple exponential decay. The boundary conditions for the exchange regime are therefore fixed by the theory: $Q_D < Q < \kappa$, which implies that the critical non-exponential relaxation can only be observed in a precise temperature and Q range and for ferromagnetic systems with low enough values of $Q_D$.

Independent renormalization Group (RG) [9] and Mode Coupling (MC) [5] calculations predict similar shapes for the spectral function at the exchange controlled regime at $Q > Q_D$. These predictions, however, were never tested thoroughly. One reason is that in neutron scattering experiments the finite resolution blurs out the magnetic relaxation spectra. Triple-axis neutron experiments (see [10] and references therein) focussed on constant-energy Q-scans which displayed a broad maximum the position of which was compared with the theoretical calculations. In this letter, we present the first detailed analysis of the shape of the relaxation function in a model 3D Heisenberg ferromagnet by taking advantage of the recent progress in the field of high resolution Neutron Spin-Echo (NSE) spectroscopy.

The RG theory expresses the spectral shape function $F(Q, \omega)$ as the real part of a complex function of the scaling variable $\kappa/Q$ and the scaled frequency $\omega/\omega_c$ , with the help of an $\epsilon = 6 - D$ expansion [9] and three parameters $k, a$ (for the critical behavior) and $b$ (for the cross-over to the hydrodynamic regime):

$$F(Q, \omega) = \frac{2}{\omega_c} \Re \left[ \left( (Z(x)\Pi_1(x, iu))^{-1} - i \frac{\omega}{\omega_c} \right)^{-1} \right]$$  
(1)



The function $\Pi_1(x, iu)$ is the self-energy of the dynamic susceptibility , approximated by:

$$\Pi_1(x, iu) = \left( 1 + bx^2 \right)^{\frac{-4}{1 + bx^2}} - i a u$$  
(2)

(where $u = \omega/\omega_c = Z(x)/X(x)$ for clarity). $Z(x)$ is essentially a scaling function, approximated by:

$$Z(x) = \frac{1 + bx^2}{1 - k \times \arctan \left( a \frac{1 + bx^2}{1 + bx^2} \right)}$$  
(3)

and $X(x)$ is the $\chi(Q)$ susceptibility modelled by a Lorentzian $1/(1 + x^2)$ in the Ornstein-Zernicke approximation. The scaling function describing the crossover with $x$ from the critical behavior to the diffusive one in the hydrodynamic regime is given by:

$$\Omega(x) = \frac{\omega_c(Q, T)}{\omega_c(Q, T_c)} = \frac{\tau_c(Q, T_c)}{\tau_c(Q, T)} = \frac{1}{X(x) X(0)}$$  
(4)

For a better comparison with the NSE experimental results, a analytical form of the intermediate Fourier transform of the spectral function [11] has been proposed:

$$I_{RG}(Q, t) = e^{-a(x) \frac{t}{\tau_c}} \left( \cos(b(x) \frac{t}{\tau_c}) + \sin(b(x) \frac{t}{\tau_c}) \right)$$  
(5)

with

$$a(x) = \frac{3.23 + 3.26x + 4.195x^2}{1 + 0.28x^{1/2} + 3.84x^{3/2}} \quad b(x) = \frac{2.93}{1 + 2.1x}$$  
(6)

and $\tau_c(Q, T) \propto \Omega(x) \tau_0 Q^{-2}$. We will use this convenient parametrization of the RG theory to reduce our NSE experimental data, at the critical temperature $T_C$ ($\Omega(x) = 1$) and above ($\Omega(x) \neq 1$).

$CdCr_2S_4$ is a normal spinel, with all $Cr^{3+}$ ions having a magnetic moment of spin only type ($S=\frac{3}{2}$) with a very low magnetic anisotropy. $CdCr_2S_4$ is a semiconductor and undergoes a ferromagnetic transition below $T_C = 84.8 \text{ K}$. The magnetic interactions are positive between the six nearest neighbors ($J_1/k_B = 13.25 \text{ K}$), negligible between the six next nearest neighbors and negative ($J_3/k_B = -0.915 \text{ K}$) between the twelve third nearest neighbors. These interactions lead to a very small dipolar wave-vector of $Q_D = 0.51 \text{ nm}^{-1}$, making this compound an ideal candidate for the study of the exchange governed critical dynamics. Our comprehensive investigation of the static critical properties ([12] and references therein), determined the critical exponents $\beta = 0.33 \pm 0.03$, $\nu = 0.70 \pm 0.03 $, $\eta = 0.065 \pm 0.030$, and $\gamma = 1.40 \pm 0.04$, which are very close to the theoretical values for the 3D Heisenberg model [13]. Therefore, $CdCr_2S_4$ is a textbook example of the 3D Heisenberg ferromagnetic class.

![Graph](image.png)

**FIG. 2:** Magnetic critical scattering at $T_C$ in $CdCr_2S_4$ , measured on the NSE spectrometer IN11 (ILL). Lines are adjustments of the RG theory of $I(Q, t)$ (see equation 5).

The measurements were performed on the neutron spin-echo (NSE) spectrometer IN11 at the Institut Laue Langevin (Grenoble, France). The polycrystalline samples were enclosed in a sealed aluminium can and placed in a standard Orange cryostat, where the temperature was controlled with a precision of $\pm 0.002 \text{ K}$ in the
1.5 – 300 K range. The transition temperature $T_C$ was determined from the maximum of the diffused intensity at small $Q$ in unpolarized mode, with correction of the offset due to the Fisher exponent $\eta$ and the finite $Q$ value. The NSE spectra were recorded with a 0.45 nm neutron wavelength in the standard paramagnetic setup with a 2D multidetector whose pixels were divided in three constant-$Q$ stripes. Separate measurements of the direct beam polarization made sure that any spurious depolarization effects from the sample were avoided. The NSE spectra were measured for $Q$, polarization effects from the sample were avoided. The direct beam polarization made sure that any spurious decay of the characteristic relaxation time $\tau$ at $T = T_C$. The adjustments of the parameter $\nu = 0.7 \pm 0.03$, $z = 2.47 \pm 0.02$ , and critical parameters: transition temperature $T_C = 84.84 \pm 0.004$ K and amplitudes $\kappa_0 = 3.61 \text{ nm}^{-1}$ and $\tau_0 = 0.02813 \text{ ns/nm}^{-2}$. This exhaustive agreement between experiment and theory is new and in some sense surprising. Previous attempts to test the critical dynamic theory with neutron triple axis spectroscopy [10, 15] adjusted the parameters $a$, $k$, and $b$ to the measured scaling function $\Omega(Q)$ over an extended $x$ range, leading to large negative values for the parameter $a$. These induce a broad overdamped maximum in $F(Q, \omega)$ (figure 5) which is definitely not compatible with experiments and our high resolution NSE.

FIG. 3: Dynamical scaling of the magnetic critical scattering at $T_C$ in $\text{CdCr}_2\text{S}_4$ for $Q \geq Q_D$. The red straight line displays the original RG model (equation 5), the blue dash line a modified RG model [15] and the green dash-dotted line a simple exponential decay of the magnetic correlations.

At $T_C$, the adjustments of the $I_{RG}(Q, T)$ parametrization to the NSE curves led to the power law decay (figure 1) of the characteristic relaxation time $\tau_c = \tau_0 Q^{-z}$ with $z = 2.47$ and $\tau_0 = 0.02813 \text{ ns/nm}^{-2}$. Figure 3 shows that all spectra collapse in a master curve when plotted against the scaling variable $t/\tau_0 Q^{-z}$. The decay of the relaxation is faster than an exponential and is surprisingly well accounted for by the original RG calculation, even though it is only a first order $\epsilon$-expansion. A modified RG model [15] is less convincing. We will comment on these modified RG versions later in the discussion.

As mentioned previously, we went beyond the trivial verification of the dynamical scaling at $T_C$, where $\kappa = \kappa/Q$ at $x = 0.49$ (upper figure) and at $x = 0.77$ (the lower figure). The blue solid lines display the RG theory at the corresponding $x$ value, the red dash lines the critical shape and the green dash-dotted lines and exponential decay.

FIG. 4: Crossover of the shape function from the critical towards an exponential decay as a function of the reduced variable $x = \kappa/Q$ at $x = 0.49$ (upper figure) and at $x = 0.77$ (the lower figure). The blue solid lines display the RG theory at the corresponding $x$ value, the red dash lines the critical shape and the green dash-dotted lines and exponential decay.
experiments (figure 2) when Fourier transformed in time. Obviously, these parameter adjustments are questionable as they are done in a much too large $x$ range. In fact, only pure Iron follows the theoretical curve over such an extended range \[8\]. A self consistent reparametrisation only for the characteristic relaxation times but also for the very shape of the relaxation function, in the nanosecond and nanometer range.

Similar studies were recently performed on Ising systems showing an exponential decay on the microsecond or second timescales using Photon Correlation Spectroscopy with either X-rays \[17\] or visible light \[18\]. However, the most detailed dynamic studies were performed by NSE \[19\] on the nanosecond timescale. In spin glasses, dynamical scaling is more a question of phase space rather than reciprocal space, with stretched exponential and power law decay \[20\] over an extended time range. The agreement between the experimental results and the theoretical predictions presented in this letter goes beyond these findings. It is the first clear experimental validation of all the implications of the dynamical scaling hypothesis not only for the characteristic relaxation times but also for the 3D Heisenberg model.

Neutron Spin Echo is a unique technique for the detailed analysis of relaxation shape functions, especially magnetic ones. In NSE resolution corrections reduce to a simple division. Moreover, the paramagnetic NSE setup discards all nuclear parts of the scattering and extracts directly the critical intermediate functions, which are as exact as possible. This simple and straightforward data reduction allowed us to check the dynamical scaling on the shape of the relaxation over an extended temperature and Q range. Our NSE study shows that the RG theory gives an accurate quantitative description of the critical scattering and dynamical scaling not only at $x = 0$ but also at the crossover to the hydrodynamic regime at $x \leq 1$. At larger $x$ values, however, the theory may not be applicable without reparametrisation either due to its approximations or to deviations of the real systems from the 3D Heisenberg model.