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Dynamic Critical Neutron Scattering from a Two-Dimensional Ising System Rb_2CoF_4

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The dynamical behavior of the two-dimensional Ising-like antiferromagnet Rb_2CoF_4, which is expected to provide a real system within the $n = 1$, $d = 2$, universality class, has been investigated near $T_N = 102.96$ K with high-resolution inelastic neutron scattering. Measurement of the characteristic frequency of the longitudinal susceptibility fluctuations yields a dynamic critical exponent $z = 1.69 \pm 0.05$. This is close to the conventional theoretical value $z = \gamma = 1.75$.

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The theoretical value of the dynamic critical exponent, z, for the two-dimensional (2D) Ising system ($n=1$, $d=2$) has attracted considerable attention over the past few years. The results of various approaches to its calculation using two models, the time-dependent Ginzburg-Landau (TDGL) model, and the single spin-flip kinetic-Ising (KI) or Glauber model, have recently been reviewed by Mazenko and Valls.[1] In this Letter we report the results of the first direct measurement of the dynamic critical exponent for rubidium cobalt fluoride, Rb_2CoF_4, using high-resolution neutron inelastic scattering techniques. This compound exhibits static magnetic behavior[2,3] which follows very closely the prediction of Onsager's exact results for the 2D Ising antiferromagnet,[4] and its dynamical behavior is expected to fall within the $n=1$, $d=2$ dynamic universality class.

Rb_2CoF_4 has the K_2NiF_4 crystal structure, $I4/mmm$, in which the Co^{2+} ions, with effective spin $S=0.5$, are arranged with separation of $a_0=4.12$ Å (at 110 K) in simple planar square arrays perpendicular to the fourfold c axis. The planes of spins are 6.80 Å apart (at 110 K) and are separated by two layers of RbF ions. The two-dimensional behavior of Rb_2CoF_4 arises both because the intraplanar exchange interaction between the spins is much stronger than the interplanar interaction, and because the interplanar interaction cancels in the mean-field approximation. The spins order antiferromagnetically along $\pm[001]$ within the layers at $T_N = 102.96 \pm 0.01$ K, at which temperature the weak interplanar interaction gives rise to full three-dimensional (3D) order. Two domains are present below T_N, and give rise to magnetic Bragg peaks at positions indicated in Fig. 1.[2] Above T_N the short-range order gives rise to diffuse rods of magnetic neutron scattering along [001] as shown. The experimental static critical exponents, $\nu_c = 0.99 \pm 0.04$, $\eta = 0.2 \pm 0.1$, $\gamma = 1.67 \pm 0.09$, $\beta = 0.115 \pm 0.016$, and $\alpha = 0.016 \pm 0.051$,[2,3,5] are within the errors equal to those calculated for the 2D Ising antiferromagnet: $\nu_c = 1$, $\eta = 0.25$, $\nu = 1.75$, $\beta = 0.125$, and $\alpha = 0$ (i.e., logarithmic divergence)[4]. However, measurement of the spin-wave energies within the planes has revealed that the transverse mode does exhibit some dispersion, from 6.17 to 7.5 THz at 6 K, and renormalizes only slightly as the temperature is raised to above T_N.[6]

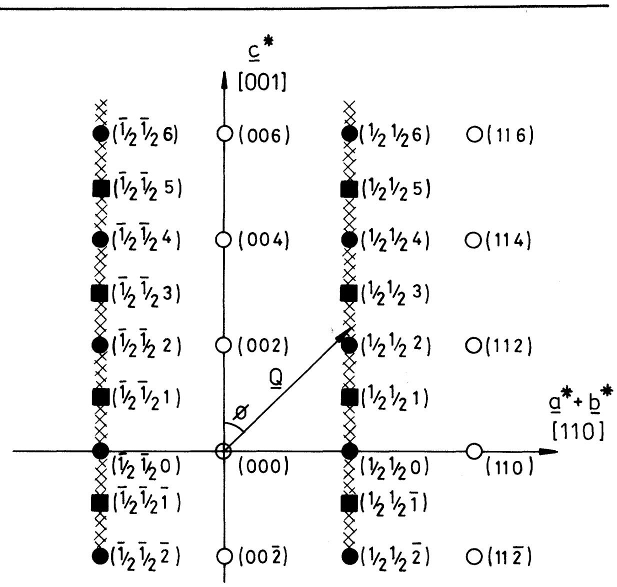

FIG. 1. The reciprocal lattice of Rb_2CoF_4. Nuclear points: open circles; magnetic points: domain 1, solid circles; domain 2, solid squares; cross hatch: diffuse magnetic rods.

The data at 6 K are fitted well by a spin Hamiltonian of the form

$$\mathcal{H} = \sum_{i,\delta} [I S_i^z S_{i+\delta}^z + J(S_i^x S_{i+\delta}^x + S_i^y S_{i+\delta}^y)], \tag{1}$$

with $S = 0.5$, $I = 1.87 \pm 0.01$ THz, and $J = 1.03 \pm 0.04$ THz. There is thus a sizable XY interaction between the spins, of 0.55 the magnitude of the Ising term, which flips the spins in this real system and gives rise to the dynamical critical scattering observed from the longitudinal susceptibility fluctuations. Nevertheless the high anisotropy, and consequent high spin-wave energies at and above T_N, will place this system well within the anisotropic region.[7-9] Indeed the spin-wave gap energy at T_N, of $\simeq 270 k_B$, is much higher than $k_B T_N$.[6]

The sample, $18 \times 12 \times 3$ mm^3, was mounted with the $[1\bar{1}0]$ direction perpendicular to the scattering plane in a standard cryostat. The temperature was controlled to ± 0.01 K and measured with a platinum resistance thermometer. All the measurements reported here were taken using the IN12 cold-neutron triple-axis spectrometer at the Institut Laue-Langevin, Grenoble. We define $\vec{Q} = \vec{k}_i - \vec{k}_f$ and $h\nu = E_i - E_f$ as the scattering vector and energy transfer, where $\vec{k}_i$ ($\vec{k}_f$) and E_i (E_f) are the initial (final) neutron wave vector and energy, respectively, and we shall denote the reduced wave vector in the 2D plane as $\vec{q} = \vec{Q} - \vec{\tau}$, where $\vec{\tau}$ is a point on the 2D reciprocal-lattice rod. Measurements were made using fixed incident neutrons with three wavelengths $\lambda_i = 6.042$, 5.236, and 4.189 Å, giving measured overall energy resolution [full width at half maximum (FWHM)] of 0.0056 ± 0.0006, 0.0104 ± 0.0010, and 0.0250 ± 0.0024 THz, respectively.

In general the scattering cross section $\sigma^D(\vec{Q}, \nu)$ will contain diffuse contributions from spin fluctuations which are both longitudinal and transverse to the c axis, and[10]

$$\sigma^D(\vec{Q}, \nu) = A|f(\vec{Q})|^2 \exp(-2W_{\vec{Q}})[(\sin^2\varphi) S^{\parallel}(\vec{Q}, \nu) + (1 + \cos^2\varphi) S^{\perp}(\vec{Q}, \nu)]. \tag{2}$$

Here φ is the angle of $\vec{Q}$ to the c axis, $f(\vec{Q})$ the magnetic form factor, $W_{\vec{Q}}$ the Debye-Waller factor, and A a constant. Because of the high anisotropy only the longitudinal contribution $S^{\parallel}(\vec{Q}, \nu)$ is expected to contribute to the observed scattering. It is given by[10]

$$S^{\parallel}(\vec{Q}, \nu) = \frac{\hbar}{\pi} \frac{1}{g^2 \mu_B^2} \frac{1}{1 - \exp(-h\nu/k_B T)} \operatorname{Im} \chi^{\parallel}(\vec{Q}, \nu) = \frac{k_B T}{g^2 \mu_B^2} \chi^{\parallel}(\vec{Q}) \frac{h\nu/k_B T}{1 - \exp(-h\nu/k_B T)} F^{\parallel}(\vec{Q}, \nu) \ldots, \tag{3}$$

where $\chi^{\parallel}(\vec{Q}, \nu)$ and $\chi^{\parallel}(\vec{Q})$ are the longitudinal dynamic and static susceptibility, $F^{\parallel}(\vec{Q}, \nu)$ is the shape function, k_B is the Boltzmann constant, and μ_B is the Bohr magneton. If a single relaxation rate is assumed, then

$$F^{\parallel}(\vec{Q},\nu)=\frac{1}{2\pi^2}\frac{\Gamma_{\parallel}(\vec{Q})}{\Gamma^2{}_{\parallel}(\vec{Q})+\nu^2},$$

and the line shape is Lorentzian centered on zero energy transfer with FWHM $=2\Gamma_{\parallel}(\vec{Q})$. $\Gamma_{\parallel}(\vec{Q})$ is the characteristic frequency[7] of the critical longitudinal fluctuations, which goes to zero at $T=T_N$. $\Gamma_{\parallel}(\vec{Q})=\kappa^z\Omega(q/\kappa)=q^z\tilde{\Omega}(q/\kappa)$, where $\Omega(q/\kappa)$ and $\tilde{\Omega}(q/\kappa)$ are universal scaling functions homogeneous in q/κ. κ is the inverse correlation length in the layer varying as $\kappa=t^{\nu}c$, where $t=(T-T_N)/T_N$, and $\Omega(q/\kappa)\to$ const as $q=0$. The dynamic critical exponent z may therefore be determined either by the κ dependence of $\Gamma_{\parallel}(\vec{Q})$ at $q=0$, or the q dependence of $\Gamma_{\parallel}(\vec{Q})$ at $\kappa=0$ (T_N).

The scattering intensity, $I(\vec{Q}_0,\nu_0)$, observed at a setting $(\vec{Q}_0,\nu_0)$ of a triple-axis spectrometer operated in the constant-k_i mode is given by the convolution integral

$$I(\vec{Q}_0,\nu_0)=\int R(\vec{Q}-\vec{Q}_0,\nu-\nu_0,\vec{Q}_0,\nu_0)\sigma(\vec{Q},\nu)d\vec{Q}\,d\nu, \quad (4)$$

where $\sigma(\vec{Q},\nu)$ is the cross section and $R(\vec{x})$ is the resolution function[11] with normalization R_0 and Gaussian dependence on $\vec{x}=(\vec{Q}-\vec{Q}_0,\nu-\nu_0)$, $R(\vec{x})=R_0\exp(-\vec{x}\cdot\vec{M}\cdot\vec{x})$. The resolution matrix $\vec{M}$ was measured carefully at each incident wave vector by a series of scans through the (002) nuclear Bragg peak.

The Néel temperature was determined by monitoring both the temperature dependence of the (0.5, 0.5, 0) magnetic Bragg peak, and the intensity of the 2D critical scattering at $h\nu=0$ at several points on the diffuse rod $(0.5, 0.5, Q_c*)$. Both methods gave the same value for $T_N=102.96\pm0.01$ K.

Measurement of the longitudinal critical scattering was made by using scans of energy transfer at three constant-$\vec{Q}$ points on the 2D reciprocal-lattice rod, $q=0$: (0.5, 0.5, 0.1), (0.5, 0.5, 0.3), and (0.5, 0.5, 2.3). Data were taken at eleven temperatures above T_N, and the q dependence of the scattering was measured at T_N at five values of q, at points $(0.5+q, 0.5+q, 0.3)$. Because of the very narrow energy widths, the extraction of $\Gamma_{\parallel}(\vec{Q})$ from the raw data required a careful convolution procedure as well as an accurate knowledge of the resolution matrix. Correction was made for a small contribution to the measured intensity from the incoherent scattering of the sample and from the general background. The theoretical expression for the intensity, Eqs. (2)–(4) with $S^{\perp}(\vec{Q},\nu)=0$, was then fitted to the data by the method of least squares using the Harwell routine FITSQW to determine values for the overall scale factor and the characteristic frequency $\Gamma_{\parallel}(\vec{Q})$ in Eq. (3). $\chi^{\parallel}(\vec{Q})$ was fixed at the value found from experiment[3]: $\sim(\kappa^2+q^2)^{-1+\eta/2}$ with $\eta=0.2$ and $\kappa=0.362t^{0.99}$, and an approximate form factor $f(\vec{Q})$ was used.[12] The fits of the Lorentzian line shape were good, with the usual criterion parameter χ^2 generally lying between 1 and 2.5.

The values of $\Gamma_{\parallel}(\vec{Q})$ obtained as a function of t at $q=0$, and as a function of q at $t=0$, are plotted in Figs. 2 and 3. There is excellent agreement between the values obtained with different k_i, and between the values obtained at different points in reciprocal space, indicating the validity of the analysis. Values for the static susceptibility $\chi^{\parallel}(\vec{\tau})$ determined from Eq. (3) by integrating the intensity over $h\nu$ were found for the three values of $\vec{Q}$. The fact that they agree with each other within the experimental error at each temperature confirms that the transverse contributions were negligible. The temperature variation of $\chi^{\parallel}(\vec{\tau})$ is found to be consistent with $\gamma=1.67\pm0.09$ determined from quasistatic data,[3] and the variation of $\chi^{\parallel}(\vec{Q})$ with q is consistent with $\eta=0.2\pm0.1$. It is seen from Fig. 2 that the meas-

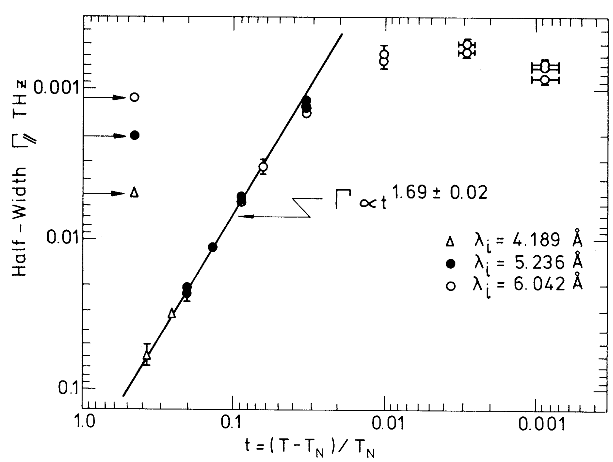

FIG. 2. Variation of characteristic frequency of longitudinal fluctuations in Rb_2CoF_4 with reduced temperature, determined at three points in reciprocal space with $q=0$. Data taken at each incident neutron wavelength are denoted by the symbol indicated. The horizontal arrows denote values of $\Gamma_{\parallel}$ equal to 0.4 of the overall energy resolution half width at half maximum at the corresponding wavelengths.

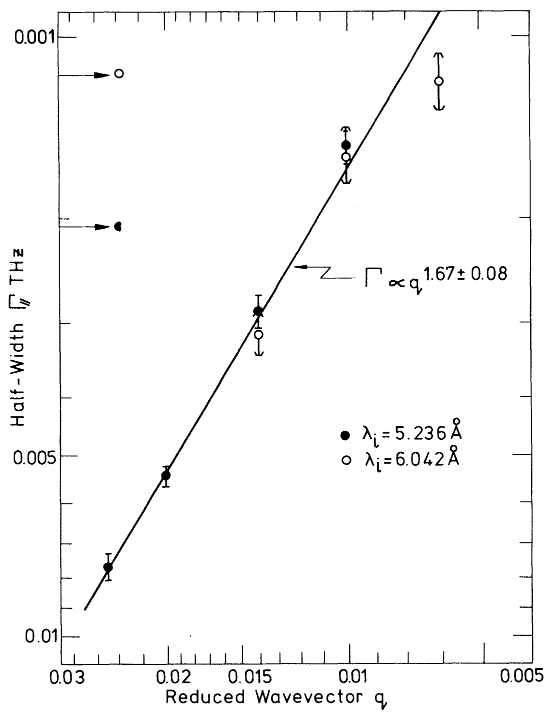

FIG. 3. Variation of characteristic frequency of longitudinal fluctuations in Rb_2CoF_4 with wave vector q at T_N. The incident neutron wavelength is denoted by the symbol indicated; the horizontal arrows are related to the energy resolution as in Fig. 2.

ured values of $\Gamma_{\parallel}(\vec{Q})$ become constant very close to T_N. As these data correspond to $\Gamma_{\parallel}$ equal to 0.1 of the overall energy resolution FWHM it is felt that they represent the limit of the experimental accuracy.

From the slopes of the lines in Figs. 2 and 3, which represent the best fit of a power-law variation, one obtains (i) in the regime $0.03 < t < 0.4$

$$\Gamma_{\parallel}(\vec{\tau}) = (0.325 \pm 0.013) t^{1.69 \pm 0.02}$$

$$= (1.84 \pm 0.27) \kappa^{1.71 \pm 0.07},$$

using the experimental variation of κ with t, and (ii) for $t = 0 \pm 0.0003$, $\Gamma_{\parallel}(\vec{Q}) = (3.6 \pm 1.2) q^{1.67 \pm 0.08}$. These indices agree with each other well within the experimental error, and their average value $z = 1.69 \pm 0.05$ is taken as the final result of the measurements.

There have been no previous direct measurements of z made on a 2D Ising-like system, but data are available for the variation of NMR linewidth and acoustic attenuation near T_N in Rb_2CoF_4, from which indirect estimates may be made. One may reanalyze the NMR data of Bucci and Guidi[13] and Bucci, Guidi, and Vignali[14] on the ^{87}Rb and ^{19}F resonance linewidth $\Delta\nu$ using the scaling relation[15] $n = \nu_c(2 - d - \eta + z)$ where $\Delta\nu \simeq t^{-n}$. Although their values of $n = 1.40 \pm 0.05$ and 1.52 ± 0.03, from the two nuclei, respectively, differ by more than the quoted error, the average value of these exponents gives $n = 1.46 \pm 0.07$, and enables one to deduce $z = 1.71 \pm 0.07$ using the theoretical values of ν_c and η, or $z = 1.67 \pm 0.13$ using the experimental values. These results are consistent with present direct and more accurate measurement which does not rely on a scaling relation. The value of $z = 1.21 \pm 0.10$ deduced from the critical variation of ultrasonic attenuation[16] is much lower. These data were taken in a region of temperature closer to T_N, but assumptions in decoupling the four-spin correlation function which may not be fully justified were made in evaluating z. A large number of attempts to calculate z have been made using the TDGL or KI models giving results which vary over a wide range, between 1.4 and 2.2, as summarized by Mazenko and Valls.[1] These authors have pointed out that even if a particular model gives accurate static critical parameters the dynamics may not be well accounted for, and that the precise value of z is not yet conclusively determined by theory. They give evidence for the possibility of regimes of dynamic scaling, with an asymptotic dynamic critical region much narrower than the asymptotic static critical region. In this asymptotic region, very close to T_N, the value of z is difficult to calculate, but away from this region several results indicate that one has dynamic scaling with a conventional value of $z = \gamma = 1.75$. The present value for Rb_2CoF_4, $z = 1.69 \pm 0.05$, lies very close to the measured $\gamma = 1.67 \pm 0.09$ and nearly encompasses the theoretical value, $\gamma = 1.75$, within the estimated error bars. It therefore strongly supports a conventional-theory value of the dynamic critical exponent.

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