

## Critical spin fluctuations in the two-dimensional antiferromagnet $\text{KFeF}_4$ : A Mössbauer study

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The critical spin fluctuations in the two-dimensional antiferromagnet  $\text{KFeF}_4$  were investigated above  $T_N = 136.15$  K in detail by means of  $^{57}\text{Fe}$  Mössbauer spectroscopy. Evidence is given that the critical dynamics of  $\text{KFeF}_4$  falls within the dynamic universality class  $(d, n) = (2, 1)$ . The dynamic critical exponent  $z$  was deduced from the longitudinal spin-autocorrelation time in the reduced temperature range  $4 \times 10^{-4} < t < 10^{-1}$ . It is shown that the dynamic critical behavior of  $\text{KFeF}_4$  is in agreement with conventional theory ( $z = 1.75$ ) with  $z = 1.77(5)$  down to at least  $t \approx 5 \times 10^{-3}$ . However, closer to  $T_N$  our data seem to indicate a small deviation from conventional behavior with  $z = 1.54(6)$ . Possible reasons for this deviation are discussed.

### I. INTRODUCTION

The static critical behavior of simple magnetic systems with short-range interactions is well understood. In this case the static critical behavior only depends on the lattice dimension  $d$  and the spin dimension  $n$ . On the other hand, the *dynamic* critical behavior of such systems is more complex. For a given static universality class  $(d, n)$  the critical dynamics depends in addition on conservation laws and Poisson bracket relations among the order parameter and the conserved densities.<sup>1</sup> Consequently, dynamic critical exponents cannot in general be expressed in terms of static exponents only.<sup>1,2</sup>

A theoretical problem of current interest in the field of critical dynamics is the evaluation of the dynamic critical exponent  $z$  for two-dimensional (2D) models belonging to the dynamic universality class  $(d, n) = (2, 1)$ , where the nonconserved order parameter is the only slow mode (model *A*).<sup>1,2</sup> In this context, two models which are be-

lieved to belong to this universality class have been studied in detail, namely, the time-dependent Ginzburg-Landau model and the single-spin-flip kinetic Ising or Glauber model.<sup>2</sup> In a review, Mazenko and Valls<sup>2</sup> have shown that all current theoretical methods to treat these models lead to inconclusive values for the dynamic exponent  $z$ , although reliable values for the static exponents are obtained. The present values for  $z$  range between 1.4 and 2.2, depending on the method involved. It has been suggested<sup>2</sup> that this discrepancy is due to the existence of an asymptotic dynamic critical region much narrower than the corresponding static region. Outside this region, theory is consistent with a conventional value of  $z = \gamma/\nu = 1.75$ , where  $\gamma$  and  $\nu$  are the static critical exponents of the susceptibility and the correlation length, respectively.<sup>2</sup> However, at temperatures sufficiently close to the critical point  $T_c$ , the conventional theory breaks down. What the true *asymptotic* value of  $z$  is, is a question of current interest.

TABLE I. Summary of dynamic critical parameters for various 2D Ising-type antiferromagnets.

Compound	$T_N(K)$	$w$	$z$	Range of $t$	Method	Ref.
$\text{K}_2\text{CoF}_4$	107.44	1.52(3)	1.74(13) <sup>a</sup>	$10^{-1} < t < 2^b$	NMR $^{19}\text{F}$	4
$\text{Rb}_2\text{CoF}_4$	99.8	1.40(5)	1.61(13) <sup>a</sup>	$10^{-1} < t < 4 \times 10^{-1b}$	NMR $^{87}\text{Rb}$	5
	103.20(2)		1.21(10)		Ultrasonic attenuation	6
	102.96(1)		1.69(5) <sup>c</sup>	$3 \times 10^{-2} < t < 4 \times 10^{-1}$	Neutron scattering	3
$\text{KFeF}_4$	135.797(6)	0.91(5)	1.29(9)	$10^{-4} < t < 10^{-3}$	Mössbauer	7
	136.131(3)	1.36(1)	1.71(4)	$4 \times 10^{-4} < t < 10^{-1}$	Mössbauer	This work
	136.150 <sup>d</sup>	1.42(2)	1.77(5)	$5 \times 10^{-3} < t < 10^{-1}$	Mössbauer	This work
	136.150(5)	1.20(4)	1.54(6)	$4 \times 10^{-4} < t < 5 \times 10^{-3}$	Mössbauer	This work

<sup>a</sup>Estimated from  $w$  as explained in Ref. 3.

<sup>b</sup>Estimated from plots in the corresponding references.

<sup>c</sup>Average value obtained at  $T_N$  and in the quoted temperature range as explained in Ref. 3.

<sup>d</sup>Fitted with fixed  $T_N = 136.15$  K.

So far only a few experiments to measure the dynamic exponent  $z$  for the universality class  $(d,n)=(2,1)$  have been performed. All experiments were done on 2D Ising-type antiferromagnets, which are believed to be good representatives of this universality class.<sup>3</sup> Various results are summarized in Table I and are discussed in Sec. III. There are some inconsistencies in the values for  $z$  determined by different techniques. Recently, Hutchings *et al.*<sup>3</sup> have investigated the critical dynamics of the 2D Ising-type antiferromagnet  $\text{Rb}_2\text{CoF}_4$  by high-resolution inelastic neutron scattering. They found a value of  $z=1.69(5)$  which is close to the conventional value, whereas earlier ultrasonic attenuation measurements by Suzuki *et al.*<sup>6</sup> yielded a considerably smaller value  $z=1.21(10)$  (Table I). A similar nonconventional value  $z=1.29(9)$  was previously found by our group for the 2D antiferromagnet  $\text{KFeF}_4$  using the Mössbauer technique.<sup>7</sup> Note that neutron scattering<sup>3</sup> allows a direct measurement of  $z$ , whereas crucial assumptions must be made to deduce  $z$  from ultrasonic attenuation<sup>6</sup> and Mössbauer<sup>7</sup> experiments.

$\text{KFeF}_4$  exhibits a static critical behavior with a critical exponent  $\beta=0.151(3)$  which is close to the exact value  $\beta=0.125$  for the 2D Ising model.<sup>7</sup> Therefore we believe that this compound also belongs to the dynamic universality class under consideration. Stimulated by the results of Hutchings *et al.*<sup>3</sup> on  $\text{Rb}_2\text{CoF}_4$ , we investigated the critical spin dynamics in  $\text{KFeF}_4$  by means of  $^{57}\text{Fe}$  Mössbauer spectroscopy in more detail.

## II. THEORETICAL BACKGROUND

Well above the critical temperature  $T_c$  of a magnetic system electronic spin fluctuations are too fast to be observed by the Mössbauer effect. As the temperature approaches  $T_c$ , however, the characteristic frequency of the critical spin fluctuations goes to zero (critical slowing down). Generally, this phenomenon gives rise to relaxation effects in the Mössbauer spectra.

Here we summarize only enough of the theory of critical dynamics to understand our results. For a detailed description of the theory we refer to Refs. 8 and 7. Fluctuations of the spin component  $S_\alpha$  ( $\alpha=x,y,z$ ) are described in terms of the space-time spin-correlation function

$$G^{\alpha\alpha}(\vec{r},t) = \langle S_\alpha(\vec{r},t) S_\alpha(0,0) \rangle / [S(S+1)/3], \quad (1)$$

or its Fourier transform, the dynamic structure factor  $S^{\alpha\alpha}(\vec{q},\omega)$ . In a Mössbauer experiment, one measures the spin-autocorrelation time  $\tau_c^\alpha$  ( $\alpha=x,y,z$ ) defined by the time integral<sup>8,7</sup>

$$\tau_c^\alpha = \frac{1}{2} \int_{-\infty}^{+\infty} dt G^{\alpha\alpha}(0,t). \quad (2)$$

It is more convenient to express this integral in terms of the dynamic structure factor  $S^{\alpha\alpha}(\vec{q},\omega)$ :

$$\tau_c^\alpha \propto \int_{v_q} d^d q S^{\alpha\alpha}(\vec{q},\omega=0), \quad (3)$$

where  $v_q$  is the Brillouin-zone volume. By using the dynamic scaling form for  $S^{\alpha\alpha}(\vec{q},0)$  (Ref. 1), an evaluation of the integral in Eq. (3) near  $T_c$  is straightforward, yielding<sup>8</sup>

$$\tau_c^\alpha \propto t^{-w}, \quad (4)$$

with

$$w = \nu(z+2-d-\eta). \quad (5)$$

Here  $t=(T-T_c)/T_c$  is the reduced temperature,  $d$  is the dimension of the system,  $\nu$  and  $\eta$  are static critical exponents, and  $z$  is the dynamic critical exponent. Equation (4) implies that at  $T_c$  the spin-autocorrelation time  $\tau_c^\alpha$  diverges with an exponent  $w$  (critical slowing down). In an experiment one measures  $w$ , from which the dynamic exponent  $z$  may be obtained according to Eq. (5), provided the static exponents  $\nu$  and  $\eta$  are known. Using the static scaling relations,  $z$  may be expressed in terms of other static exponents (e.g.,  $\beta$ ,  $\gamma$ , and  $\nu$ ):<sup>7</sup>

$$z = d(w+2\beta)/(\gamma+2\beta), \quad (6a)$$

$$z = (w+2\beta)/\nu. \quad (6b)$$

These relations are most convenient here since  $\beta$  has previously been evaluated by the same technique.<sup>7</sup>

In order to extract the spin-autocorrelation time  $\tau_c^\alpha$  from the Mössbauer spectra, an appropriate relaxation theory is needed. In this work we adopt the results of Bradford and Marshall<sup>9</sup> who have calculated the  $^{57}\text{Fe}$  Mössbauer line shape in the limit of fast electronic relaxation using perturbation theory. In contrast to our previous work,<sup>7</sup> where the spin relaxation was assumed to be isotropic, we consider here a more general case. The hyperfine interaction  $V(t)$  is taken to have axial symmetry:

$$V(t) = A_{\parallel} I_z S_z(t) + A_{\perp} [I_x S_x(t) + I_y S_y(t)], \quad (7)$$

where  $A_{\parallel}$  and  $A_{\perp}$  are the hyperfine coupling constants parallel and perpendicular to the  $c$  axis, respectively. It is further assumed that the spin-autocorrelation function  $G^{\alpha\alpha}(0,t)$  in Eq. (2) has an exponential form:

$$G^{\alpha\alpha}(0,t) = \delta_{\alpha z} \exp(-|t|/\tau_c^{\parallel}) + (\delta_{\alpha x} + \delta_{\alpha y}) \exp(-|t|/\tau_c^{\perp}), \quad (8)$$

with different correlation times  $\tau_c^{\parallel}$  and  $\tau_c^{\perp}$ , parallel and perpendicular to the  $z$  direction, respectively. In the case of a single-crystal absorber with the principal component  $V_{zz}$  of the axially symmetric electric field gradient (EFG) tensor parallel to the direction of the  $\gamma$  rays (cf. Sec. III), the relaxation spectrum may be approximated by two Lorentzians:<sup>9</sup>

$$A(\omega) \propto \frac{(\Gamma_0 + \Delta\Gamma_1)/2}{(\omega - \Delta/2)^2 + [(\Gamma_0 + \Delta\Gamma_1)/2]^2} + \frac{1}{3} \frac{(\Gamma_0 + \Delta\Gamma_2)/2}{(\omega + \Delta/2)^2 + [(\Gamma_0 + \Delta\Gamma_2)/2]^2}, \quad (9)$$

where  $\Gamma_0$  is the natural linewidth [full width at half maximum (FWHM)], and  $\Delta$  is the quadrupole splitting. The corresponding line broadenings  $\Delta\Gamma_{1,2}$  due to the relaxation are

$$\Delta\Gamma_1 = \frac{S(S+1)}{6\pi^2} [(A_{\parallel}^0 - 3A_{\parallel}^*)^2 \tau_c^{\parallel} + 2(A_{\perp}^0 + 3A_{\perp}^*)^2 \tau_c^{\perp}], \quad (10a)$$

$$\Delta\Gamma_2 = \frac{S(S+1)}{6\hbar^2} [(A_{\parallel}^0 + A_{\parallel}^*)^2 \tau_c^{\parallel} + 2(A_{\perp}^{0^2} + 7A_{\perp}^{*2}) \tau_c^{\perp}]. \quad (10b)$$

Here  $A_{\alpha}^0$  and  $A_{\alpha}^*$  denote the hyperfine coupling constants of the ground and the excited states, respectively. The validity of Eqs. (9) and (10) is discussed in Sec. III.

### III. RESULTS AND DISCUSSION

The structural and magnetic properties of the layered antiferromagnet  $\text{KFeF}_4$  are well described in Refs. 10 and 7. Simply speaking,  $\text{KFeF}_4$  consists of slightly tilted and distorted  $\text{FeF}_6$  octahedra, separated by nonmagnetic layers of  $\text{K}^+$  ions. Below the Néel temperature  $T_N = 136.15$  K the magnetic moments ( $\text{Fe}^{3+}, S = \frac{5}{2}$ ) are aligned along the  $c$  axis (perpendicular to the layers). Moreover, the inter-layer exchange interaction  $J'$  is much weaker than the intralayer interaction  $J$  ( $|J'/J| \approx 10^{-4}$ ), giving rise to the quasi-two-dimensional magnetic properties of  $\text{KFeF}_4$ .

The spectra were taken on a single crystal of  $\text{KFeF}_4$  with the direction of the  $\gamma$  rays perpendicular to the magnetic layers. The crystals were grown by the flux-growth method as described in detail in Ref. 11. In order to reduce finite thickness effects and temperature gradients in the sample, a rather thin and small crystal ( $60 \mu\text{m} \times 5 \text{mm}^2$ ) was used, in contrast to the larger sample used in the previous work.<sup>7</sup> Special attention was paid to avoid temperature gradients and mechanical strains across the sample by mounting the crystal stress-free in a massive copper sample holder with additional radiation shields. The details of the cryostat and the temperature control system are given in Ref. 7. The long-term temperature stability of the sample was better than  $\pm 3$  mK/24 h. Independent cooling and heating runs during several months gave the same results within experimental errors. The velocity of the drive was constantly monitored and calibrated with a laser interferometer.

A spectrum of  $\text{KFeF}_4$  taken at 4.2 K is shown in Fig. 1. It clearly illustrates the high quality of our sample. The corresponding hyperfine parameters as obtained by a least-squares fit (solid line in Fig. 1) are identical with the previous results.<sup>7</sup> From the low-temperature spectra it is known<sup>7</sup> that the hyperfine field  $H$  is perpendicular to the layers and that the principal component  $V_{zz}$  of the axial

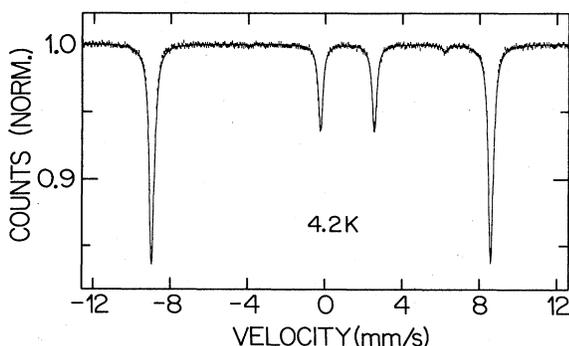


FIG. 1. Low-temperature Mössbauer spectrum of single-crystal  $\text{KFeF}_4$ .

EFG tensor is negative and almost parallel to  $H$  (the actual angle is about  $12^\circ$ ).

Typical quadrupole spectra taken at various temperatures above  $T_N = 136.15$  K are represented in Fig. 2. Notice the obvious broadening of the lines as  $T_N$  is approached. Similar relaxation spectra were previously observed by Ito and Horiike<sup>12</sup> for the 2D Ising-type antiferromagnet  $\text{Rb}_2\text{CoF}_4$ :<sup>57</sup>Fe. The broadening of the left line ( $\frac{3}{2} \rightarrow \frac{1}{2}$ ) is much more pronounced than that of the right line ( $\frac{1}{2} \rightarrow \frac{1}{2}$ ) (see also Fig. 3). Since the principal component  $V_{zz}$  of the axial EFG tensor is almost parallel to the direction of the  $\gamma$  rays, we may approximate the spectra with the line shape given by Eqs. (9) and (10). Thus the spectra were fitted to two Lorentzians, taking corrections for the finite absorber thickness into account. The solid curves in Fig. 2 represent the fitted line shape. The temperature dependence of the linewidths  $\Gamma_{1,2} = \Gamma_0 + \Delta\Gamma_{1,2}$  (FWHM) is shown in Fig. 3. The average value  $\Gamma_0 = 0.198(2)$  mm/s as obtained from several spectra well above  $T_N$  is consistent with the experimental source linewidth  $\Gamma_0 = 0.1974(13)$  mm/s for an infinitely thin absorber. The latter value of  $\Gamma_0$  was used to extract the line broadenings  $\Delta\Gamma_{1,2}$  from the measured linewidths  $\Gamma_{1,2}$ .

A quantity  $p$  which measures the degree of anisotropy of the spin relaxation may be derived from the line broadenings using Eq. (10):

$$p = (A_{\perp} / A_{\parallel})^2 (\tau_c^{\perp} / \tau_c^{\parallel}). \quad (11)$$

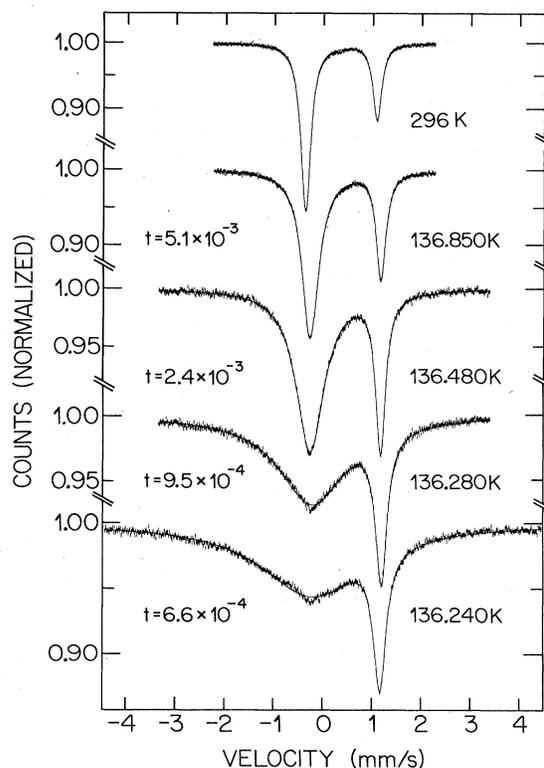


FIG. 2. Mössbauer spectra of single-crystal  $\text{KFeF}_4$  taken in the critical region above  $T_N = 136.15$  K.  $t$  denotes the reduced temperature. For comparison, a room-temperature spectrum is shown at the top.

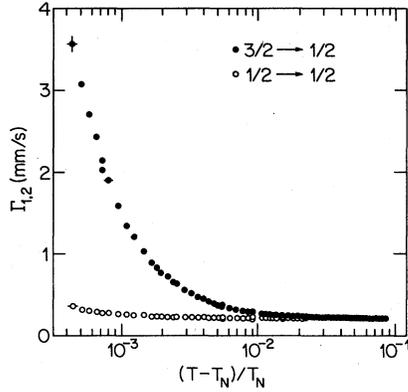


FIG. 3. Mössbauer linewidths  $\Gamma_1$  (closed circles) and  $\Gamma_2$  (open circles) as a function of reduced temperature.

The temperature dependence of  $p$  stems from that of the correlation times  $\tau_c^{\parallel}$  and  $\tau_c^{\perp}$ . For isotropic spin relaxation  $p=1$ , whereas  $p=0$  for pure longitudinal spin relaxation. A plot of  $p$  extracted from the data is shown in Fig. 4 as a function of reduced temperature. For a weakly anisotropic Heisenberg system<sup>7</sup> such as  $\text{KFeF}_4$  ( $S=\frac{5}{2}$ ) the spin fluctuations are expected to be isotropic ( $p=1$ ) well above  $T_N$ . As the temperature approaches  $T_N$ , however, the spin fluctuations become anisotropic and the relaxation spectra are dominated by the longitudinal fluctuations ( $p \simeq 0$ ) as seen in Fig. 4. This important observation further justifies our assumption that  $\text{KFeF}_4$  belongs to the universality class under consideration (cf. Sec. I). It is known from neutron scattering experiments<sup>13,3</sup> on 2D antiferromagnets that the critical scattering is determined only by the longitudinal component  $S^{\parallel}(\vec{q}, \omega)$  of the dynamic structure factor. Therefore in a Mössbauer experiment the longitudinal spin-autocorrelation time  $\tau_c^{\parallel}$  defined in Eq. (3) should reflect the true critical behavior.

The temperature dependence of the experimental  $\tau_c^{\parallel}$  as obtained from Eq. (10) is shown in Fig. 5. Notice that in the reduced temperature region investigated,  $\tau_c^{\parallel}$  increases by 3 orders of magnitude from  $10^{-12}$  to  $10^{-9}$  s as  $T_N$  is approached from above. The exponent  $w$  defined in Eqs. (4) and (5) was determined by fitting the data to the power law

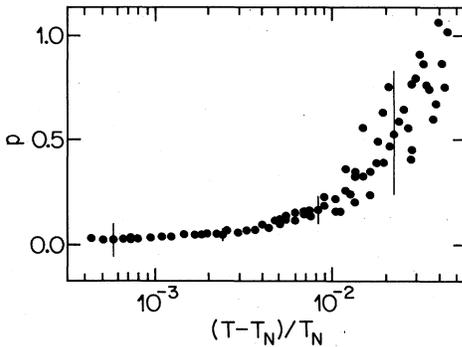


FIG. 4. Anisotropy parameter  $p$  of spin fluctuations versus reduced temperature. The definition of  $p$  is given in the text [cf. Eq. (11)].

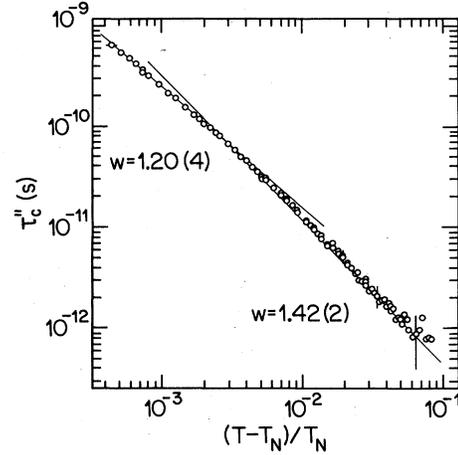


FIG. 5. Longitudinal spin-autocorrelation time  $\tau_c^{\parallel}$  as a function of reduced temperature. The slopes of the straight lines determine the critical exponents  $w=1.20(4)$  and  $1.42(2)$  obtained below and above  $t \simeq 5 \times 10^{-3}$ , respectively (Table I).

$$\tau_c^{\parallel} = \tau_0 (T/T_N - 1)^{-w}, \quad (12)$$

with  $\tau_0$ ,  $T_N$ , and  $w$  as adjustable parameters. In order to systematically investigate the behavior of  $w$  near  $T_N$ , the following fitting procedure was adopted:<sup>14,7</sup> A series of weighted least-squares fits to Eq. (12) were performed by successively omitting data points on the high-temperature side until only the 15 data points closest to  $T_N$  remained (further reduction of the number of data points lead to large uncertainties in  $w$ ). The variation of  $w$  as a function of the maximum reduced temperature  $t_{\max}$  is shown in Fig. 6. For  $t_{\max} > 3 \times 10^{-2}$ ,  $w$  is almost constant. However, below  $t_{\max} \simeq 3 \times 10^{-2}$ ,  $w$  decreases continuously, and at  $t_{\max} \simeq 5 \times 10^{-3}$  a distinct change in  $w$  to a lower and nearly constant value is observed, accompanied by a remarkable reduction in  $\chi^2$ . The dashed lines in Fig. 6 correspond to the best values  $w=1.36(1)$  and  $1.20(4)$  as obtained in the reduced temperature ranges  $4 \times 10^{-4} < t < 10^{-1}$  and  $4 \times 10^{-4} < t < 5 \times 10^{-3}$ , respectively. A best fit to the data in the more restricted range  $5 \times 10^{-3} < t < 10^{-1}$  with  $T_N$  fixed at  $T_N=136.15$  K yields  $w=1.42(2)$ . This value is somewhat larger than the global value  $w=1.36(1)$  (see Table I). The slopes of the straight lines in Fig. 5 are determined by the values of  $w$

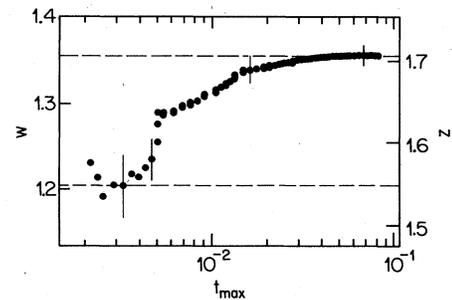


FIG. 6. Variation of the exponent  $w$  with maximum reduced temperature  $t_{\max}$  as obtained from fits to the power law in Eq. (12). The right-hand scale shows the corresponding values of the dynamic exponent  $z$ .

below and above  $t \approx 5 \times 10^{-3}$ .

The dynamic exponent  $z$  was calculated from the values of  $w$  listed in Table I using Eqs. (6a) and (6b). The static exponent  $\beta=0.151(3)$  was previously measured using the same apparatus and crystals of the same origin.<sup>7</sup> Since the static exponents  $\gamma$  and  $\nu$  are not known for  $\text{KFeF}_4$ , we determined  $z$  with assumptions similar to those made in our previous work,<sup>7</sup> i.e.,  $\gamma+2\beta=1.95(5)$  and  $\nu=0.95(6)$  (Ref. 15). With these assumptions and the measured value of  $\beta$  one obtains a value of  $z=\gamma/\nu=1.74(12)$ . The large error in  $z$  is due to the large uncertainty in  $\nu$ . This value is in agreement with the theoretical conventional value of  $z=1.75$ , indicating that the assumptions concerning the static exponents are reasonable. The experimental values of  $z$  quoted in Table I are weighted averages of  $z$  as obtained from Eqs. (6a) and (6b). In the following the present values of  $z$  for  $\text{KFeF}_4$  are compared with the previous value and with those reported for other 2D Ising-type antiferromagnets (Table I). The most striking result is that the value of  $z=1.71(4)$  as obtained from a best fit over the whole temperature region investigated ( $4 \times 10^{-4} < t < 10^{-1}$ ) is consistent with conventional theory ( $z=1.75$ ). This finding is in agreement with the recent high-resolution neutron scattering results of Hutchings *et al.*<sup>3</sup> for  $\text{Rb}_2\text{CoF}_4$  (Table I). Similar values of  $z$  can also be derived from earlier NMR results of Bucci *et al.*<sup>4,5</sup> for  $\text{Rb}_2\text{CoF}_4$  and  $\text{K}_2\text{CoF}_4$ . Note that the NMR measurements were taken far away from  $T_N$  (Table I). In the temperature region very close to  $T_N$  ( $4 \times 10^{-4} < t < 5 \times 10^{-3}$ ), however, the present data yield a value of  $z=1.54(6)$  which slightly deviates from conventional theory (see also Figs. 5 and 6). This value is more reliable than the previous value  $z=1.29(9)$ , obtained in a similar reduced temperature range,<sup>7</sup> for the following reasons. (i) In the previous work<sup>7</sup> it was assumed that the spin fluctuations in this weakly anisotropic 2D Heisenberg antiferromagnet are isotropic. From the present work it is evident that this is not the case for temperatures close to  $T_N$  (see Fig. 4). For  $t < 10^{-2}$  the relaxation spectra are dominated by the longitudinal spin fluctuations. This is due to a small Ising-type anisotropy in the exchange interaction perpendicular to the magnetic layers, giving rise to a 2D Ising-type critical behavior<sup>16</sup> which is also reflected in the static critical exponent  $\beta=0.151(3)$  measured previously.<sup>7</sup> For comparison, note that for  $\text{Rb}_2\text{CoF}_4$  a similar nonconventional value of  $z=1.21(10)$  was deduced from ultrasonic attenuation measurements<sup>6</sup> also requiring important assumptions.<sup>3</sup> (ii) The present measurements were taken on a much smaller and thinner crystal in order to avoid serious temperature gradients and to keep the corrections for the finite absorber thickness as small as possible.

A principal question arises whether the observed deviation from conventional theory is physically real or just an artifact of the relaxation model adopted. The Bradford-Marshall theory<sup>9</sup> is a reasonable approach, since the correlation times  $\tau_c$  are sufficiently short ( $\tau_c < 7 \times 10^{-10}$  s) for Eqs. (9) and (10) to be valid, in first order. Support for the model is offered by the fact that the spectra are rather well described over virtually the whole temperature range by two Lorentzians, as shown in Fig. 2. Notice, however,

that for  $t < 10^{-3}$  small deviations from a Lorentzian line shape are apparent, which probably arise from nondiagonal terms in the hyperfine interaction due to the small angle between the principal component  $V_{zz}$  of the EFG tensor and the hyperfine field  $H$ . A further assumption is that the spin-correlation function  $G^{\alpha\alpha}(0,t)$  has an exponential form [Eq. (8)]. This is not the case for critical fluctuations where the correlation function has a long tail.<sup>17</sup> It may be shown,<sup>17</sup> however, that the Bradford-Marshall theory<sup>9</sup> is still valid for a general correlation function if the correlation times  $\tau_c^\alpha$  in Eq. (10) are defined by the time integral in Eq. (2). Consequently, for the model to hold the correlation function must decay sufficiently rapidly, since in practice the integration limits in Eq. (2) are finite; this may be problematic for a critical correlation function with a long tail. Thus it may not be excluded that the observed kink in  $\tau_c^{\parallel}$  close to  $T_N$  (Fig. 5) is an artificial effect. However, for  $t > 5 \times 10^{-3}$  the spin-correlation times are sufficiently short ( $\tau_c^{\parallel} < 3 \times 10^{-11}$  s) for our assumptions to hold rigorously. This is also evident from Fig. 2, where for  $t > 5 \times 10^{-3}$  no deviation from a Lorentzian line shape is observed. As a result, in the restricted reduced temperature range,  $5 \times 10^{-3} < t < 10^{-1}$ , a true value of  $z=1.77(5)$  was found (Table I) which is in excellent agreement with the conventional value  $z=1.75$  predicted by theory.

#### IV. CONCLUSIONS

The critical spin dynamics in the 2D-layered antiferromagnet  $\text{KFeF}_4$  was investigated above  $T_N$  in some detail by means of Mössbauer spectroscopy. The main conclusions of this work may be summarized as follows.

The critical spin fluctuations in  $\text{KFeF}_4$  are found to be anisotropic; the longitudinal component reflecting the critical behavior is perpendicular to the magnetic layers. A best fit to the longitudinal spin-autocorrelation time over the whole reduced temperature range,  $4 \times 10^{-4} < t < 10^{-1}$ , yields a dynamic critical exponent  $z=1.71(4)$ . This value is consistent with conventional theory ( $z=1.75$ ) and is in agreement with the experimental value  $z=1.69(5)$  for the 2D Ising-type antiferromagnet  $\text{Rb}_2\text{CoF}_4$  measured by high-resolution inelastic neutron scattering.<sup>3</sup>

At  $t \approx 5 \times 10^{-3}$  a change in  $z$  from the conventional value to a smaller value  $z=1.54(6)$  is observed (Figs. 5 and 6) which appears to indicate the onset of nonconventional behavior. This experimental value lies within the range  $z=1.4-2.2$  predicted by current theory.<sup>2</sup> However, one cannot guarantee that this is not an artifact of the relaxation model used.

For  $t > 5 \times 10^{-3}$ , where the spectra show no deviations from Bradford-Marshall theory<sup>9</sup> (solid lines in Fig. 2), a value of  $z=1.77(5)$  was obtained. This value of  $z$  is even closer to the conventional theoretical value  $z=1.75$  than that deduced in the whole reduced temperature range. Thus we conclude that conventional theory for  $\text{KFeF}_4$  is essentially valid down to at least  $t \approx 5 \times 10^{-3}$ . Our results are consistent with the theoretical prediction of Mazenko and Valls<sup>2</sup> that for the dynamic universality class  $(d,n)=(2,1)$ , model *A*, the asymptotic dynamic critical re-

gion is much narrower than the corresponding static region. For comparison, the static critical region for  $\text{KFeF}_4$  extends up to almost  $t \approx 10^{-1}$  (Ref. 7). For all these reasons we conclude that the dynamic critical behavior of the 2D antiferromagnet  $\text{KFeF}_4$  falls most likely within this particular universality class.

In this paper we demonstrated that the Mössbauer effect provides a complementary method to other experimental techniques (e.g., neutron scattering) to study critical spin dynamics in magnetic systems. However, there is a common problem with all current experimental

methods: It appears to be very difficult to determine a reliable *asymptotic* value of  $z$  for this dynamic universality class since experiments very close to the critical point ( $t < 10^{-4}$ ) are difficult to perform and to interpret.

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- <sup>1</sup>P. C. Hohenberg and B. I. Halperin, *Rev. Mod. Phys.* **49**, 435 (1977).
- <sup>2</sup>G. F. Mazenko and O. T. Valls, *Phys. Rev. B* **24**, 1419 (1981).
- <sup>3</sup>M. T. Hutchings, H. Ikeda, and E. Janke, *Phys. Rev. Lett.* **49**, 386 (1982).
- <sup>4</sup>C. Bucci and G. Guidi, *J. Phys. (Paris) Colloq.* **32**, C1-887 (1971).
- <sup>5</sup>C. Bucci, G. Guidi, and C. Vignali, *Solid State Commun.* **10**, 803 (1972).
- <sup>6</sup>M. Suzuki, K. Kato, and H. Ikeda, *J. Phys. Soc. Jpn.* **49**, 514 (1980).
- <sup>7</sup>H. Keller and I. M. Savić, *Phys. Rev. B* **28**, 2638 (1983), and references therein.
- <sup>8</sup>C. Hohenemser, L. Chow, and R. M. Suter, *Phys. Rev. B* **26**, 5056 (1982), and references therein.
- <sup>9</sup>E. Bradford and W. Marshall, *Proc. Phys. Soc. London* **87**, 731 (1966). See also A. M. Afanas'ev and V. D. Gorobchenko, *Zh. Eksp. Teor. Fiz.* **66**, 1406 (1974) [*Sov. Phys.—JETP* **39**, 690 (1974)].
- <sup>10</sup>M. Eibschütz, G. R. Davidson, and H. J. Guggenheim, *Phys. Rev. B* **9**, 3885 (1974); G. Heger and R. Geller, *Phys. Status Solidi B* **53**, 227 (1972).
- <sup>11</sup>B. M. Wanklyn, *J. Mater. Sci.* **10**, 1487 (1975).
- <sup>12</sup>A. Ito and M. Horiike, *J. Phys. (Paris) Colloq.* **40**, C2-290 (1979).
- <sup>13</sup>R. J. Birgeneau, J. Als-Nielsen, and G. Shirane, *Phys. Rev. B* **16**, 280 (1977).
- <sup>14</sup>R. M. Suter and C. Hohenemser, *J. Appl. Phys.* **50**, 1814 (1979).
- <sup>15</sup>This value of  $\nu$  is an average of experimental values for various 2D antiferromagnets and is assumed to be a more confident estimate than  $\nu=0.9(1)$  used in Ref. 7.
- <sup>16</sup>K. Binder and D. P. Landau, *Phys. Rev. B* **13**, 1140 (1976).
- <sup>17</sup>A. M. Gottlieb and C. Hohenemser, *Phys. Rev. Lett.* **31**, 1222 (1973); M. A. Kobeissi, R. Suter, A. M. Gottlieb, and C. Hohenemser, *Phys. Rev. B* **11**, 2455 (1975).