

manner that the usual Josephson supercurrent is a general phenomenon occurring whenever two superconductors are weakly coupled.

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¹⁴Because of the narrow temperature range over which exponential behavior is exhibited (see Fig. 3), it is possible to represent our data as being exponentially dependent on T with only a slight decrease in the goodness of fit. However, fitting our data with such an expression for I_c leaves Fig. 2 essentially unchanged.

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Experimental Study of the Two-Dimensional Ising Antiferromagnet Rb_2CoF_4

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The critical behavior of Rb_2CoF_4 has been studied by means of neutron scattering. In its long-range order dependence below $T_N = 103$ K the material was found to behave as an almost ideal two-dimensional Ising system, with critical parameters $B = 1.16 \pm 0.03$ and $\beta = 0.119 \pm 0.008$. A novel phenomenon was discovered below T_N : The three-dimensional stacking of the antiferromagnetic planes is sensitive to the heat treatment given at T_N . Above T_N the inverse correlation length and the staggered susceptibilities were found to obey power laws with exponents $\nu = 0.89 \pm 0.1$ and $\gamma = 1.34 \pm 0.22$.

We report here briefly on a neutron scattering study of the critical behavior of the two-dimensional Ising-like antiferromagnet Rb_2CoF_4 .

The material has the same crystal structure as K_2NiF_4 , a compound thoroughly studied by Birgeneau and co-workers.¹ Its two-dimensional character is due to the geometrical structure consisting effectively of antiferromagnetic planes 13.7 Å apart. However, as pointed out for K_2NiF_4 ,¹ long-range two-dimensional order below the Néel point T_N leads automatically to ordering also in the third dimension due to the weak but finite interplanar coupling; whereas the Ni compound is expected to be a good Heisenberg system due to the orbital singlet ground state of Ni^{2+} in the fluoride octahedra, the Co compound will be more Ising-like because of the doublet ground state of Co^{2+} , which introduces a strong magnetic anisotropy.² In fact Breed, Gilijamse, and Miedema² estimate for Rb_2CoF_4 from their sus-

ceptibility measurements a ratio of 4.35 between the longitudinal and the transverse components of the effective exchange interaction.

Figure 1 shows the observed long-range order parameter $\sigma(T)$ versus temperature as obtained from the magnetic Bragg peaks. For comparison, $\sigma(T)$ for three other compounds and the theoretical curve for the two-dimensional Ising model are also shown. The latter is seen to reproduce the Rb_2CoF_4 data exceedingly well; our data thus represent the first direct experimental corroboration of the celebrated Onsager solution.⁴ In terms of critical parameters the observations near T_N can be represented by

$$\sigma(T) = B(-\epsilon)^\beta, \quad (1)$$

$$\epsilon \equiv (T - T_N)/T_N. \quad (2)$$

B and β take the values 1.16 ± 0.03 and 0.119 ± 0.008 , respectively, as obtained by a least-

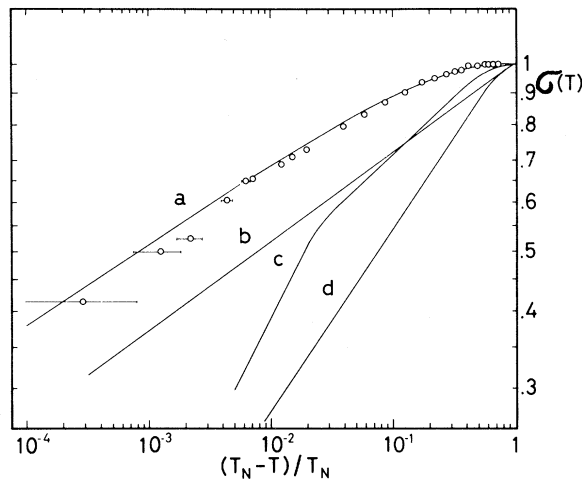


FIG. 1. Reduced sublattice magnetization $\sigma(T)$ for Rb_2CoF_4 as determined from one series of neutron-diffraction measurements. The curves shown are *a*, the theoretical curve for a two-dimensional $S = \frac{1}{2}$ Ising system; *b*, neutron measurements for K_2NiF_4 (Ref. 1); *c*, for Rb_2FeF_4 (Ref. 1); *d*, NMR measurements (Ref. 3) for the three-dimensional MnF_2 .

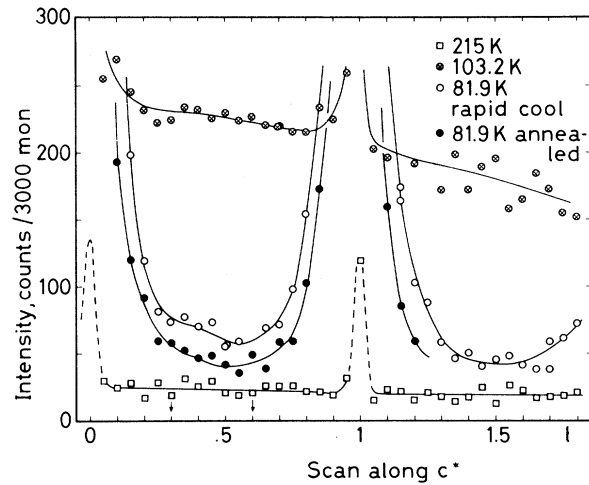


FIG. 2. Scans along the ridge for four different cases: Far above T_N (215 K), just above T_N (103.2 K), and below T_N (81.9 K) for both a rapidly cooled and annealed crystal. The arrows show the points on the ridge at which the temperature variation of Fig. 3 has been studied. The Bragg scattering below T_N occurs at integral l (0, 1). Some higher-order contamination is seen to remain at 215 K.

squares fitting to 44 observations for $T > 90$ K. The respective theoretical values are 1.22 and 0.125. The fitted value of T_N is 103.025 ± 0.01 K.

Near and above T_N the two-dimensionality was observed by the occurrence of neutron intensity ridges in the reciprocal space through the magnetic reciprocal-lattice points, as for K_2NiF_4 .¹ It was found that some remnant of the ridges in fact remained at low temperatures too as seen in Fig. 2, their widths and intensities being essentially unaltered below $0.9T_N$. In addition, the width of the Bragg peaks was found to be strongly heat-treatment dependent. Quick cooling through T_N gave rise to broader magnetic Bragg peaks and also increased the ridge intensity, indicating that the ordering among the planes is not completely long range. From the peak widths one could deduce that a cooling rate of 2 K/min gave rise to an average domain size of 50 c -axis units, whereas careful annealing could double or triple this number. Full long-range order seemed to be impossible to establish with our samples. We attribute the occurrence of these metastable states to the very abrupt variation of $\sigma(T)$ just below T_N , which makes the phase transition almost discontinuous.

Above T_N the width and intensity dependence of the ridge were studied, as seen in Fig. 3. No energy width was observed to the scattering at

any temperature. The data have therefore been analyzed in the quasielastic approximation.¹ From the observed widths we deduce the temperature dependence of κ , the inverse correlation length:

$$\kappa = C\epsilon^\nu, \tag{3}$$

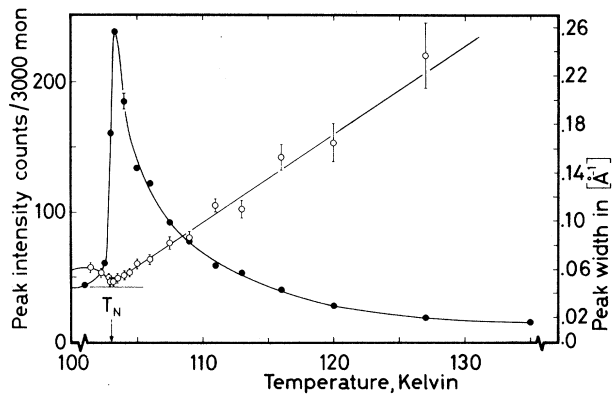


FIG. 3. Observed temperature dependence of the two-dimensional ridge, showing peak intensity (solid circles), which measures the staggered susceptibility, and peak width across the ridge (open circles), which determines the inverse correlation length κ . The width has a minimum at T_N , increases somewhat below T_N , and reaches a constant value at lower temperature as indicated by the horizontal line.

with $C = 0.283 \pm 0.044 \text{ \AA}^{-1}$ and $\nu = 0.89 \pm 0.1$. The value of ν is close to the theoretical value of unity for a two-dimensional Ising system.

The power-law temperature dependence of the staggered susceptibility was found to hold over a more restricted temperature range than for κ . The exponent γ takes the value $\gamma = 1.34 \pm 0.22$, whereas theory predicts 1.75. An independent determination of the Fisher exponent η could not be made, but from the scaling law relation⁵ $\gamma = \nu(2 - \eta)$ one gets $\eta = 0.5 \pm 0.1$. This value is just the double of the theoretical 0.25.

Although the ridge intensity of Fig. 3 is seen to peak strongly near T_N , the detailed behavior very close to T_N shows that the divergency is considerably less than predicted by theory⁵ (after proper correction for the instrumental resolution). In fact the observed intensity maximum occurs about 0.2 K above T_N . Related to this is the observation of an *extra scattering component at the reciprocal-lattice points* for temperatures between T_N and $T_N + 0.2$ K. These two findings have been interpreted as an indication of a tran-

sition from two-dimensionality to three-dimensionality in the critical behavior as T_N is approached from above. Based on a semiquantitative model for the transition a ratio between the interplanar to the intraplanar coupling constants of about 2×10^{-3} could be estimated. A more detailed account of the work will be published elsewhere.⁶

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Magnetolectric Determination of the Pressure-Induced T_N Shift in Cr_2O_3 †

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We report a new technique for studying the effect of uniaxial stress and hydrostatic pressure on the Néel temperature of antiferromagnets. Using magnetolectric susceptibility measurements, we studied the shift in the Néel temperature T_N of Cr_2O_3 caused by an externally applied uniaxial stress (\vec{a} and \vec{c} axis) or hydrostatic pressure p . Contrary to previously reported results, $(dT_N/dp)_c$ was approximately 70% greater than $(dT_N/dp)_a$.

We report on a new technique based upon the utilization of magnetolectric (ME) susceptibility measurements¹ to determine the dependence of the Néel point T_N on an applied uniaxial stress or hydrostatic pressure. Since the ME effect occurs only in the ordered phase and is known¹ to disappear abruptly at the phase transition, ME susceptibility measurements are a uniquely sensitive method of determining T_N as a function of some external parameter.

Our initial study was carried out on single crystals of antiferromagnetic Cr_2O_3 . In general, the ME susceptibility α_{ij} arises from a term in the free energy of the form

$$- \alpha_{ij} E_i H_j, \quad (1)$$

where E_i and H_j are components of the local electric and magnetic fields, and summation over repeated indices is implied. For the case of antiferromagnetic Cr_2O_3 with symmetry $3'm'$, the