Critical behavior of two-dimensional Rb_2CoF_4 as observed by linear birefringence

P. Nordblad,* D. P. Belanger,† A. R. King, and V. Jaccarino
Department of Physics, University of California, Santa Barbara, California 93106

H. Ikeda
Department of Physics, Ochanomizu University, Ohtsuka, Bunkyo-ku, Tokyo 112, Japan
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The temperature derivative of the linear birefringence $d(\Delta n)/dT$ is used to measure the magnetic specific heat C_m of the two-dimensional (2D) Ising antiferromagnet Rb_2CoF_4. A direct comparison is made between Onsager's exact solution of the 2D Ising model and experiment. Excellent agreement was found throughout the critical region and to the lowest temperatures. Departure of experiment from theory well above T_N is attributed to short-range-order contributions arising from the partly Heisenberg character to the exchange interaction in Rb_2CoF_4.

Rb_2CoF_4 is a layered antiferromagnet in which the static[1,2] and dynamic[3,4] critical behavior have been shown to agree well with the theoretical predictions for the two-dimensional (2D) Ising model.[5] It has been shown theoretically[6] that, within the critical region, the temperature derivative of the linear birefringence $d(\Delta n)/dT$ is proportional to the magnetic specific heat C_m. Such a proportionality has been established experimentally in the three-dimensional (3D) Ising systems FeF_2 and MnF_2.[7] Specific-heat[8] and birefringence measurements[9] for K_2CoF_4 indicate that the proportionality holds for the 2D systems as well. Studies of Rb_2CoF_4 (Ref. 8) suggests the specific heat exhibits a symmetric logarithmic behavior in the critical region. However, the analysis of the behavior of C_m is hampered by the relatively large phonon contribution to the total specific heat. By way of contrast, $d(\Delta n)/dT$ is almost entirely magnetic in origin. We present birefringence data for Rb_2CoF_4 in the temperature range $40 \leq T \leq 300$ K which allows a precise comparison to be made between the measured C_m and the exact Onsager theory over a wide temperature range.

A 2-mm-thick single crystal of Rb_2CoF_4, with parallel faces cleaved perpendicular to the unique axis, was oriented with the face 45° to a laser beam at $\lambda = 632.8$ nm. A Sénarmont compensator, with 50-kHz modulation, was used to measure the effective birefringence $\Delta n'$, which is proportional to the true birefringence Δn of the crystal which would be directly observed if the unique axis were perpendicular to the laser beam.[9] The sample temperature was measured with a resolution of 100 μK at 100 K. The resolution in Δn of $\sim 10^{-8}$ was limited primarily by the optical quality of the sample.

Figure 1 shows the overall temperature dependence of $d(\Delta n')/dT$ from 40 to 300 K, where the points shown are the difference in $\Delta n'$ between consecutive data points divided by the temperature difference ΔT and are plotted at the average temperatures. The same is true for the data shown in Figs. 2 and 3. That the nonmagnetic contribution to $d(\Delta n')/dT$ is extremely small is quite evident from the data far from T_N. The behavior of C_m in the critical region is shown in Fig. 2 where $d(\Delta n')/dT$ is plotted versus $\log_{10}|t|$ and $t = (T - T_N)/T_N$. The data have been fitted to the function

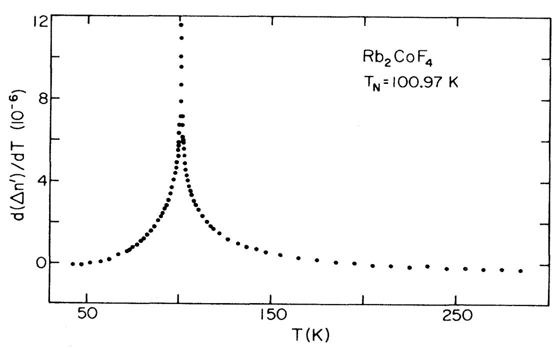

FIG. 1. $d(\Delta n')/dT$ vs T in Rb_2CoF_4.

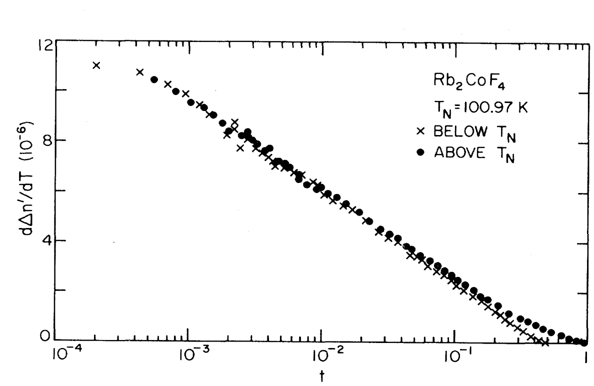

FIG. 2. $d(\Delta n')/dT$ vs $\log_{10}|t|$ in Rb_2CoF_4 showing the symmetric logarithmic divergence in the critical region $|t| < 0.1$.

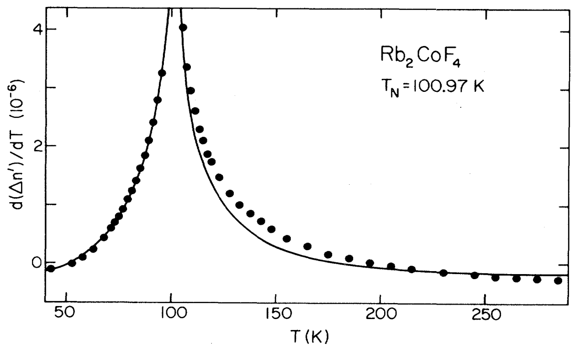

FIG. 3. $d(\Delta n')/dT$ vs T in Rb_2CoF_4. The solid line is the exact Onsager solution with amplitude adjusted to fit the data, and a constant background term $d(\Delta n')/dT = 0.16\times 10^{-6}$ subtracted.

$$\frac{d(\Delta n')}{dT} = \frac{A}{\alpha}(|t|^{-\alpha} - 1) + B + Et \qquad (1)$$

for $t > 0$ and the same expression with amplitude A' for $t < 0$. For the range $5\times 10^{-4} \le |t| \le 5\times 10^{-2}$ values of $\alpha = -0.002\pm 0.013$, $A/A' = 1.01\pm 0.01$, and $T_N = 100.97$ K were obtained. A similar fit was made over a somewhat larger temperature region, $5\times 10^{-4} \le |t| \le 0.1$, with essentially no change in the parameters. This temperature range is well within the critical region since neutron scattering results[4] have shown that the inverse correlation length $K = 0.362 t^{0.99}$. This yields $K^{-1} \sim 28$ Å or $6.7a_0$ at $t = 0.1$. A complete list of parameters is given in Table I.

Since the 2D Ising transition is the only physically realizable model for which there is an exact solution, and the data are of such high quality, it is interesting to compare experiment with theory over the entire range of temperatures, including those well outside the critical region. To do this we have expanded the vertical scale of the data in a linear plot in Fig. 3 to compare it with the complete Onsager solution for the 2D Ising system. Because the proportionality factor between the birefringence and C_m is unknown, it is treated as an adjustable parameter. As has been shown in detail for the 3D Ising systems FeF_2 and MnF_2, there is a small phonon contribution to $d(\Delta n')/dT$ which is clearly manifest in the behavior far from T_N. In Rb_2CoF_4 this contribution is apparently negative, causing $d(\Delta n')/dT$ to be negative both far above and far below T_N. To accurately estimate the lattice background to $d(\Delta n')/dT$ would require a comparison of the birefringence in Rb_2CoF_4 with that in a nonmagnetic isomorphic crystal (e.g., Rb_2ZnF_4) as was done for FeF_2 and ZnF_2, in the 3D case.[10] Since $d(\Delta n')/dT$ of the nonmagnetic Rb_2ZnF_4 is not presently available, we have simply subtracted a constant lattice contribution $[d(\Delta n')/dT = 0.16\times 10^{-6}]$ from the theory, to agree with the data below T_N.

TABLE I. Parameters obtained from fitting $d(\Delta n')/dT$ for Rb_2CoF_4 to Eq. (1).

	$0.0005 \le \|t\| \le 0.05$	$0.0005 \le \|t\| \le 0.1$
T_N	100.966 ± 0.004	100.962 ± 0.004
α	-0.002 ± 0.013	-0.003 ± 0.010
A/A'	1.010 ± 0.008	1.020 ± 0.007
$10^6 A'$	$1.56\ \pm 0.07$	$1.53\ \pm 0.10$
$10^6 B$	$-1.09\ \pm 0.17$	$-1.14\ \pm 0.24$
$10^6 E$	$0.61\ \pm 0.54$	$1.23\ \pm 0.87$

The agreement between experiment and theory is seen to be excellent for $T < T_N$, but the data for $T > T_N$ lie significantly above the theory, especially in the region around $1.5T_N$. We believe this to result from the non-negligible Heisenberg-type character of the Hamiltonian. The latter, via the effects of short-range-order, contributes a broad maximum to C_m.

It is interesting to examine the effects that the Heisenberg-type contribution to the exchange Hamiltonian have on the isomorphic 2D magnetic system. This is illustrated very clearly in $d(\Delta n')/dT$ in the series K_2NiF_4, Rb_2NiF_4, Ba_2NiF_4, and K_2CoF_4,[11] where the Heisenberg contribution decreases monotonically. In K_2NiF_4 and Rb_2NiF_4, the Ising anomaly at T_N is a tiny peak on the much larger, broad Heisenberg background. In Ba_2NiF_4 both features are equally prominent and in K_2CoF_4, the most Ising-type of the series, as in Rb_2CoF_4, the Heisenberg component is barely noticeable. While no theory exists for such a mixed Ising-Heisenberg model, either inside or outside the critical region, it is clear that the data are of sufficient quality to permit an accurate comparison with theory, when and if it appears.

In conclusion, we find the optical birefringence technique to have sufficient sensitivity and such an extremely small lattice contribution as to allow accurate comparison with the exact Onsager theory over a wide range of temperature. At the same time, the small deviations from the 2D Ising model, which are observed above T_N, are shown to be consistent with the additional weak, but finite, Heisenberg-type character of the exchange interaction in Rb_2CoF_4.

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*Permanent address: Department of Solid State Physics, University of Uppsala, Box 534, S-75121 Uppsala, Sweden.
†Present address: Department of Physics, Brookhaven National Laboratories, Upton, New York 11973.

[1]E. J. Samuelsen, Phys. Rev. Lett. 31, 936 (1973); J. Phys. Chem. Solids 35, 785 (1974).
[2]H. Ikeda, M. Suzuki, and M. T. Hutchings, J. Phys. Soc. Jpn. 46, 1153 (1979).
[3]C. Bucci, G. Guidi, and C. Vignali, Solid State Commun. 10, 803 (1972).

[4]M. T. Hutchings, H. Ikeda, and E. Janke, Phys. Rev. Lett. 49, 386 (1982).
[5]L. Onsager, Phys. Rev. 65, 117 (1944).
[6]G. A. Gehring, J. Appl. Phys. 53, 8152 (1982).
[7]I. R. Jahn, Phys. Status Solidi 57, 681 (1973); D. P. Belanger, P. Nordblad, A. R. King, V. Jaccarino, L. Lundgren, and O. Beckman, J. Magn. Magn. Mater 31-34, 1095 (1983). 1982 [J. Magn. Magn. Mater. (in press)].
[8]H. Ikeda, L. Hatta, and M. Tanaka, J. Phys. Soc. Jpn. 40, 334 (1976).
[9]K. Iio, M. Sakatani, and K. Nagata, J. Phys. Soc. Jpn. 43, 709 (1977).
[10]D. P. Belanger, Ph.D. thesis, University of California at Santa Barbara, 1981 (unpublished); D. P. Belanger, A. R. King, and V. Jaccarino (unpublished).
[11]K. Iio, M. Sakatani, and K. Nagata, J. Phys. Soc. Jpn. 45, 1567 (1978).