Critical behavior and critical endpoints of $FeCl_2 \cdot 2H_2O$ and $CoCl_2 \cdot 2H_2O$ in an applied magnetic field

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$FeCl_2 \cdot 2H_2O$ and $CoCl_2 \cdot 2H_2O$ represent metamagnets with two discontinuities in the magnetization at low temperatures. An antiferromagnetic (AF) phase at low fields is followed by a ferrimagnetic phase at intermediate fields and a paramagnetic (P) phase at high fields. These three phases meet at one point at a higher temperature. By means of neutron-diffraction experiments, we have investigated the properties of the systems in the vicinity of this point. For $FeCl_2 \cdot 2H_2O$, this point is shown to be a critical endpoint ($H_{CEP}=4.329\pm0.007$ T; $T_{CEP}=11.20\pm0.05$ K), i.e., an endpoint of a λ line of continuous transitions emanating from the Néel point and ending on a first-order transition line. In zero field, we find the critical exponent for the sublattice magnetization to be $\beta=0.30\pm0.01$; near the critical endpoint (CEP) we find $\beta_{CEP}=0.29\pm0.02$, i.e., no crossover on the whole λ line. For $CoCl_2 \cdot 2H_2O$, we find a triple point ($H_{TP}=3.972\pm0.009$ T; $T_{TP}=9.53\pm0.01$ K). The critical exponent β in zero field is observed to be $\beta_3=0.32\pm0.01$ for $\epsilon_T=(T_N-T)/T_N<6\times10^{-3}$ but $\beta_1=0.18\pm0.01$ for $6\times10^{-3}<\epsilon_T<6\times10^{-2}$; this is interpreted as crossover in effective spatial dimensionality. Under applied fields, the $d=3$ behavior disappears on the AF-P-phase boundary at $H_t=4.00\pm0.01$ T; $T_t=10.0\pm0.1$ K. Here we find an exponent $\beta_t=0.130\pm0.005$. We discuss in detail whether this may be understood as a tricritical point at (H_t,T_t) or whether the triple point (H_{TP},T_{TP}) should be characterized as a critical endpoint as in $FeCl_2 \cdot 2H_2O$, so excluding the existence of a tricritical point: The issue cannot be fully resolved.

I. INTRODUCTION

The work of Griffiths[1] (in 1970) on multicritical points has stimulated new interest on systems with such a behavior. In 1972, Riedel and Wegner presented the first renormalization-group (RG) treatment of tricritical points (TCP).[2] They found general agreement of RG tricritical exponents with those calculated by Landau theory. As an example, the exponent β_t of the order parameter $M_s \sim (H_t-H)^{\beta_t}$ should be $\beta_t=\frac{1}{4}$ at the tricritical point. This is valid only for the lattice dimensionality $d \geq 3$, but for all values of the spin dimensionality n. For $d=3$, additional logarithmic corrections are probably necessary.[3] During the last few years, many experimental studies on systems with such tricritical points have been performed, especially on metamagnetic systems such as $FeCl_2$ (Refs. 4–6) and $CsCoCl_3 \cdot 2D_2O$.[7] Detailed information on experimental results on metamagnetic compounds can be found in the literature.[8] Metamagnets are Ising systems. In applied magnetic fields parallel to the easy direction, these systems only show spin reversals but no change of the spin orientation into directions perpendicular to the easy direction, i.e., no spin-flop phases as found for weak anisotropies. Below the temperature T_t of the TCP, there are field-dependent jumps of magnetization M and sublattice magnetization M_s. At temperatures $T \geq T_t$ the phase transitions are continuous with the above-described behavior of the order parameter $M_s \sim (H_C-H)^{\beta}$.

In the following we present our results on critical behavior of the metamagnetic compounds $FeCl_2 \cdot 2H_2O$ (FC2) and $CoCl_2 \cdot 2H_2O$ (CC2). At temperatures below $T_N=21.5$ K (FC2) and $T_N=17.3$ K (CC2), both systems order in an antiferromagnetic phase (AF). By contrast to most metamagnets, both systems exhibit *two* successive jumps of the magnetization M and sublattice magnetization M_s. This holds for temperatures below a temperature T_{TP} of a triple point (H_{TP},T_{TP}). For temperatures below T_{TP}, the AF phase is followed by a ferrimagnetic phase (FI) at intermediate fields and a paramagnetic phase (P) at high fields. At the temperature T_{TP}, the FI phase disappears. $CoBr_2 \cdot 2H_2O$ (Ref. 9) (CB2) and $RbFeCl_3 \cdot 2H_2O$ (Refs. 10 and 11) represent other compounds with such behavior. Figure 1 shows the (H,T) phase diagram of CC2.[12] Our aim has been to clarify by new experiments whether there is a new kind of critical behavior, caused by the existence of the intermediate phase, or whether there is just an ordinary tricritical behavior with a tricritical point (TCP) on the AF-P phase boundary and with normal first-order phase transitions below this TCP. From molecular-field (MF) theory there exist statements about the kind of phase transitions, specifically for our compounds.[13] Therefore, the aim of our experiments was to look for differences between these MF results and experiments, so as to stimulate new theoretical work by modern RG theories and Monte Carlo methods. In Sec. II we present what is known about the

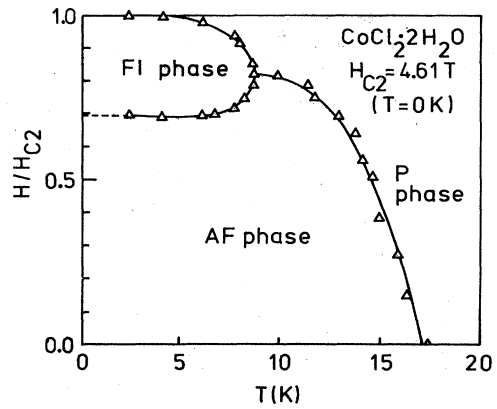

FIG. 1. $(H_{\vec{b}}, T)$ phase diagram of $CoCl_2 \cdot 2H_2O$ (from Ref. 12).

phase transitions of these compounds from the literature in theory and experiment. Sections III and IV report our new experiments: geometrical aspects and the critical behavior. In Secs. V and VI, we discuss our experiments in the framework of existing theoretical treatments in detail. Section VII concludes the paper by comparing briefly the results for the two compounds.

II. MAGNETIC PHASE TRANSITIONS IN FC2 AND CC2

FC2 and CC2 crystallize in isomorphous chemical ($C2/m$) and magnetic (P_C2_1/a and P_C2/m, respectively) structures. The individual magnetic moments are coupled in ferromagnetic chains along the $\vec{c}$ axis with an exchange interaction $J_0 > 0$. Such chains are coupled together by two exchange interactions: an antiferromagnetic interaction $J_1 < 0$ from the unit-cell corner to the $\vec{a},\vec{b}$-face center and an antiferromagnetic interaction $J_2 < 0$ along the $\vec{a}$ axis. A magnetic exchange coupling J_3 along the $\vec{b}$ axis is negligibly small because of the screening of the water molecules. The following inequality is valid: $J_0 > |J_1| > |J_2|$. In the AF phase the interaction J_1 determines the AF structure consisting of two antiparallel sublattices, one formed by the unit-cell corners and the other one formed by the $\vec{a},\vec{b}$-face centers. In FC2, the easy axis $(\vec{a})$ lies within the $(\vec{a},\vec{c})$ plane, whereas in CC2, the easy axis is along the $\vec{b}$ direction. In the FI phase, complete chains have reversed their spin directions. There results a honeycomblike ferrimagnetic structure with a unit cell trebled in the direction of the $\vec{a}$ axis and a magnetization of one third of the saturation magnetization M_S. The nature of the magnetic phases and their extension in the (H,T) plane have been extensively studied by us.[12,14–16]

The results of the MF calculations mentioned above for FC2 and CC2 can be summarized as follows:[13] The topology of the (H,T) diagram as well as the character of the phase transitions are completely determined by the constants J_0, J_1, and J_2. These constants can be calculated given the Néel temperature T_N, the paramagnetic Curie temperature Θ, and the transition fields H_{C1} and H_{C2} at 0 K as follows:[13,17,18] $2k\Theta = 2J_0 + 4J_1 + 2J_2$; $J_2/J_1 = 2(H_{C2} - H_{C1})/(2H_{C2} + H_{C1})$, and $kT_N = -4J_1 + 2J_0 + 2J_2$. In these equations, the signs of the exchange interactions J_i and the notation of the transition fields H_{C1} and H_{C2} are chosen according to Refs. 17 and 18, i.e., in contrast to Ref. 13. So far MF calculations are in qualitative agreement with the experimental phase diagrams; that of CC2 is shown in Fig. 1. Some special features of the phase diagrams, resulting from the experimental values of T_N, Θ, H_{C1}, and H_{C2} should be pointed out:[13] All phase transitions from the FI phase are of first order, i.e., discontinuous. For the temperature T_t of the TCP, one obtains $T_t = T_N + 2T_N J_1/(3J_0 + 3J_2) = T_N + 2T_N J_1/(3k\Theta - 6J_1)$. For CB2, the AF phase arches over the FI phase forming a sickle-shaped AF region near the triple point (see Fig. 4 of Ref. 13). The AF-P phase boundary is a line of continuous transitions up to the triple point because one finds $T_t < T_{TP}$. In contrast to CB2, the phase diagram of CC2 should exhibit a TCP on the AF-P phase boundary, because now one has $T_t > T_{TP}$. FC2 should behave more like CC2 than like CB2.[13] Some further considerations[19] based on MF theory should be mentioned: They indicate two unspecified multicritical points, one on the FI-P phase boundary and the other on the AF-P phase boundary.

Our previous experimental work[12,14–16] on FC2 and CC2 confirmed the phase diagram as calculated by MF theory,[13] but we found a part of the FI-P boundary with a continuous transition near the triple point. From this follows the existence of a tricritical point on the FI-P border and of a triple point in the form of a bicritical point, both in agreement with the other MF analysis.[19]

Our new experiments, which we will report in the following sections, yield everywhere a first-order phase transition for the FI-P phase boundary. This is contrary to what appears on a first inspection (see, e.g., Fig. 4 of Ref. 12), and it was not manifested until we studied the critical phenomena of both systems. Generally MF theory is not correct in details. Critical fluctuations shift the phase boundaries, and they may change the kind of multicritical behavior. For our systems, modern theoretical treatments do not exist, but we can refer to the analogous situation of spin-flop systems with bicritical points.[20] Experiments on CB2 also agree only qualitatively with MF results. For instance, the AF-P phase boundary does not extend over the FI phase; rather, it shifts below it.[9]

III. EXPERIMENTS—PHASE BOUNDARIES

Single crystals were grown by a method described elsewhere.[21] The $\vec{c}$ axis is always parallel to the main growth direction of the crystal. Single-crystal neutron-diffraction experiments were carried out on the two-axes diffractometers P49 at the FR2 reactor, Karlsruhe, and D10 at the Institut Laue-Langevin (ILL) reactor, Grenoble. The incident wavelengths were λ(P49) = 1.069 Å, λ(D10, FC2) = 2.3649 Å, and λ(D10,CC2) = 2.3630 Å. The samples ($V \simeq 2 \times 2 \times 7$ mm^3) were mounted within a 6.4-T split-pair cryomagnet of our laboratory which is equipped with a variable-temperature insert. The temperatures of the two samples were measured via a Ge-resistor thermometer and a Au/Fe-chromel thermocouple, respective-

ly. In order to study the field and temperature dependence of the magnetic scattering, we recorded about 1000 ω scans for each compound around the reciprocal-lattice points (010) for FC2 and (100) for CC2. The reflection profiles were analyzed for Bragg and Lorentz scattering by means of our computer program NLSQ.[22]

The experimental phase diagram of FC2 is shown in Fig. 2. It is in agreement with our former results.[15] Only the location of the AF-FI phase boundary differs from that which we have published previously[15] insofar as the unusual curvature below 8 K is absent. This time we located the boundary by scanning the reflections (100) and (010) right after cooling the sample from the paramagnetic state down to low temperatures at $H=0$. Only in this case, called the "first run," does the crystal exist in a virgin state, in contrast to the states of second and further runs of the magnetic field, at which the temperature between the different field runs was kept always below T_N. The new transition field is $H_{C1}=3.7$ T at $T=2$, 3, and 4 K. The previously observed AF-FI curvature with $H_{C1}=4.0$ T in this temperature range could be reproduced after the sample had been in the FI state at least once, with the temperature kept below T_N. The sweep rates of the magnetic field were 9 and 18 Oe/sec. However, the transition field was not influenced by these two different sweep rates nor by the field direction relative to the $\vec{a}$ axis. The field has to be adjusted parallel to the $\vec{c}$ axis for the reflection (100) and parallel to the $\vec{a}$ direction for the reflection (010). In the former case we converted the measured field $H_{C1}(\vec{c})$ into an effective field $H_{C1}(\vec{a})$ simply by using $H_{C1}(\vec{a})=H_{C1}(\vec{c})\cos\varphi$, in which $\cos\varphi$ represents the projection of the measured applied field onto the easy direction $\vec{a}$ with $\varphi(\vec{c},\vec{a})=32.9°$. All experiments in connection with the AF-FI phase boundary were performed with the same crystal.

We can demonstrate by the following experiment the fact that the previously observed curved phase boundary

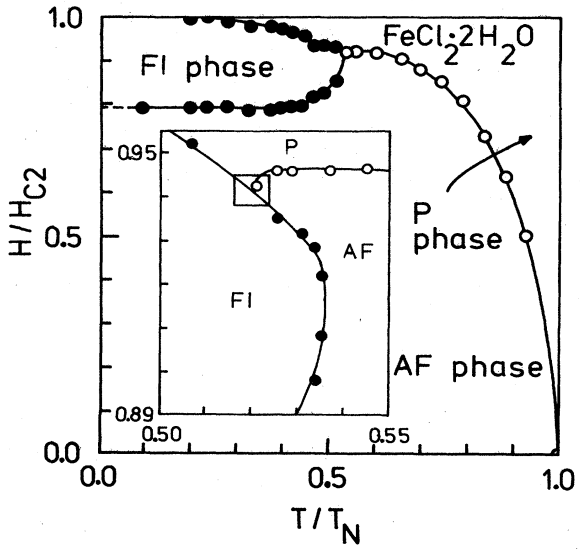

FIG. 2. $(H_{\vec{a}},T)$ phase diagram of $FeCl_2 \cdot 2H_2O$; $H_{C2}=4.6$ T ($T=0$ K); $T_N=21.5$ K; open and closed circles represent second- and first-order phase transitions.

is not stable. Coming from zero field at 2 or 3 K in a second, third, or further run, we stopped raising the field at 3.9 T and found that the AF reflections (100) and (010) disappeared after some minutes, whereas the FI reflection $(\frac{2}{3}00)$ appeared simultaneously. This demonstrates a metastable behavior, and we believe that the correct location of the AF-FI boundary is the one where we detect the first indications of a breakdown of the AF-phase reflections for sweep rates as slow as possible and in a first run.

The question arises whether there exists a homogeneous metastable magnetic structure at these temperatures and in the field region between 3.7 and 4.0 T or whether the phase boundary is simply shifted from 3.7 to 4.0 T. Concerning this question, it has been reported that a metastable phase exists below 4 K with a bulk magnetization of $\frac{1}{2}$ that of the fully aligned phase.[23] In particular, these magnetization experiments show that at 4.2 K the AF-FI transition field H_{C1} increased from the first to the second runs as we have found (Fig. 5 of Ref. 23). Second, the transition field increased with rising sweep rate, i.e., when the sweep rate of the magnetic field was increased from 20 to 60 Oe/sec (Fig. 5 of Ref. 23). Below 4 K and at 3.97 T, the above-mentioned $M_S/2$ magnetization appears, and it is seen more clearly for a second than for a first run.[24] This measured $M_S/2$ magnetization was explained[23] by a new homogeneous, metastable, ferrimagnetic "$M_S/2$-state" appearing only for sweep rates such as 60 Oe/sec and only for temperatures below 4 K and fields above 3.97 T. In summary, this means that it is not only possible to shift the AF-FI phase boundary to higher fields by various procedures, but even to produce a second ferrimagnetic phase. Some models for the magnetic structure of this metastable phase have been proposed:[23] one with doubled $\vec{a}$ or $\vec{b}$ axis and one with doubled $\vec{a}$ and $\vec{b}$ axes.

Unfortunately, it was not possible for us to increase the sweep rate of our Nb_3Sn-tape split-pair magnet to a value higher than 18 Oe/sec to study this proposed new phase. Nevertheless, in one case with our slow sweep rate of 18 Oe/sec, we could demonstrate the existence of a reflection $(\frac{1}{2}00)$ with low intensity by an ω scan at 3.85 T and 3 K. We believe that we detect a remnant of a $M_S/2$ phase. Search for a reflection $(0\frac{1}{2}0)$ was fruitless in every case. This suggests a magnetic structure for the metastable phase with a unit cell $2\vec{a},\vec{b},\vec{c}$ and excludes the other proposed model with a unit $2\vec{a},2\vec{b},\vec{c}$. Nevertheless, because of the small intensity, we cannot decide whether the reported $M_S/2$ magnetization belongs to a homogeneous, well-defined $M_S/2$ state or not. We believe that one cannot exclude a mixture of two or more phases, e.g., of the AF,FI ($M_S/3$), and P phases, existing in this region of the phase diagram for high sweep rates and resulting in a bulk $M_S/2$ magnetization. Raman experiments which can separate the different magnetic phases as in neutron diffraction but not in bulk magnetization measurements have been used to search for a $M_S/2$ state also.[25] With a sweep rate of 100 Oe/sec, no $M_S/2$ state was detected. Until now it is not clear how much time can pass from starting at zero field and moving up to a field of 3.97 T, at which the $M_S/2$ state exists, in order to have it persist

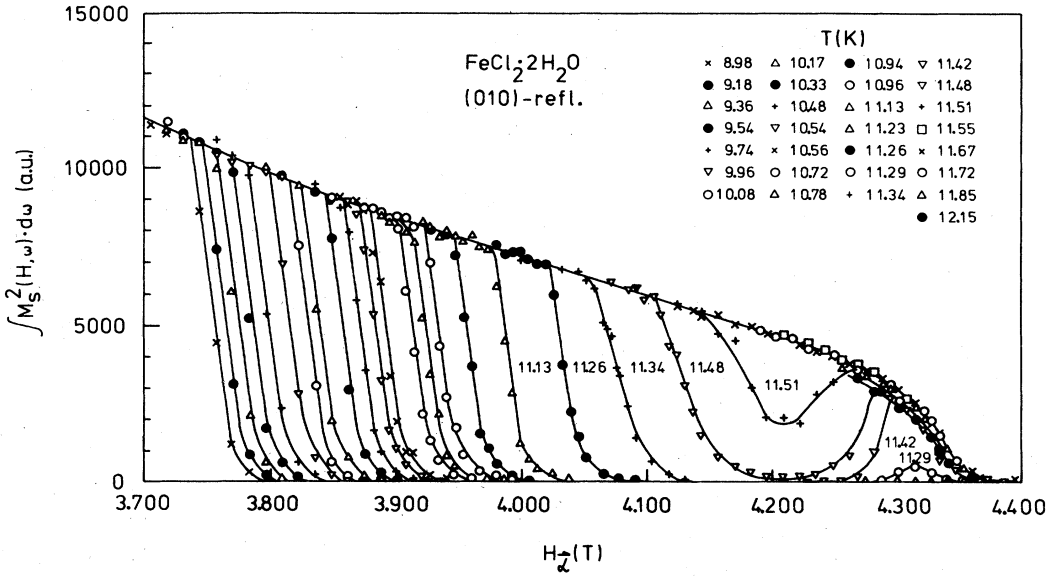

FIG. 3. Integrated intensities of the reflection (010) of $FeCl_2 \cdot 2H_2O$ as a function of the applied field $H_{\vec{a}}$ for different temperatures.

for a sufficiently long time. Therefore we believe that more experiments are necessary to confirm the existence of a homogeneous $M_S/2$ state.

Figure 3 shows the dependence of the integrated intensity of the reflection (010) on the applied field for constant temperatures for FC2. We want to emphasize the sharpness of the discontinuous jumps. They are smeared out only over a region of 0.04 T corresponding to 1%. This width is completely determined by the inhomogeneity of the magnetic field, so there is no need to perform a correction of the data for demagnetization as in cases of high demagnetization factors.[26] For example, $FeCl_2$ shows strong demagnetization effects amounting to as much as 50% (see Fig. 1 of Ref. 5). For CC2 the situation is the same as for FC2. We believe that demagnetization effects, which are the result of the formations of domains, are hindered by the intermediate FI phase. The critical lines have been located by methods we used to determine critical exponents. This will be demonstrated below.

Let us describe some geometrical aspects of the phase diagrams by means of Fig. 2 (inset) and Fig. 4 for FC2 and Fig. 5 for CC2. As can be seen, FC2 exhibits no tricritical point; rather the AF-P phase transitions are continuous up to the FI phase boundary. Therefore, the point where the AF, FI, and P phase come together represents a

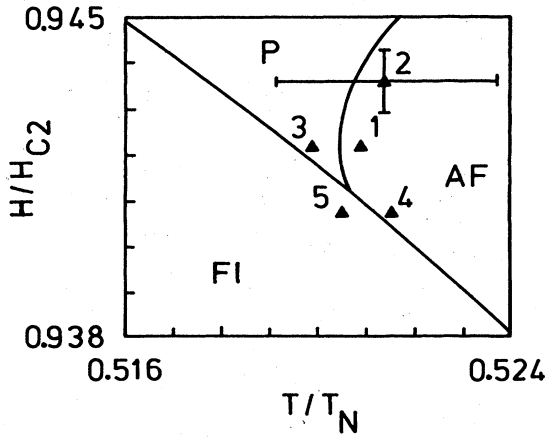

FIG. 4. Part of the $(H_{\vec{a}},T)$ phase diagram of $FeCl_2 \cdot 2H_2O$. Positions 1—5 represent the (H,T) coordinates of five ω scans, i.e., no positions of phase boundaries (see Fig. 6). The error bars at position 2 are related to the absolute accuracy of T and H.

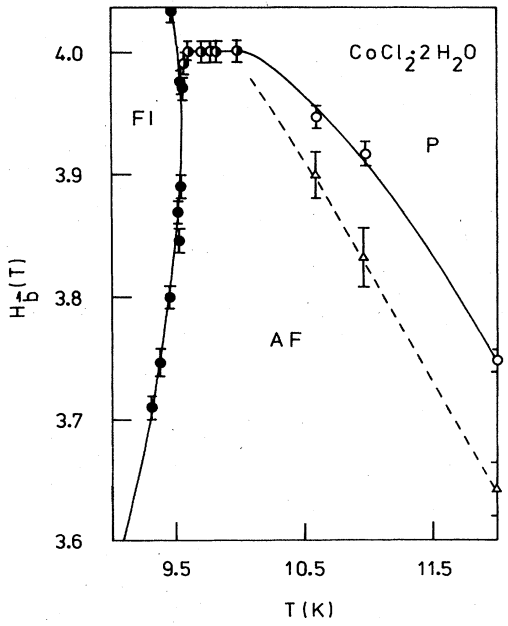

FIG. 5. Part of the $(H_{\vec{b}},T)$ phase diagram of $CoCl_2 \cdot 2H_2O$; closed and open circles represent first- and second-order transitions, respectively. At the positions with half-closed circles, a second-order transition is only verified for $\epsilon_H \geq 3\times10^{-3}$. A dimensional crossover line is marked by triangles.

critical endpoint (CEP).[27–31] In previous work, the notation "λ point" was used, e.g., for the two well-known "upper and lower λ points"[32] which exist in the (p,T) diagram of He^4. The coordinates of the CEP are $H_{CEP}=4.329\pm0.007$ T and $T_{CEP}=11.20\pm0.05$ K. Moreover, we find a sickle-shaped region arching over the FI phase as was found by MF theory for CB2 but not for FC2.[13] The sickle bends down into the CEP, apparently developing a cusp of the P phase (see Fig. 4). As mentioned in Sec. II, we believe this bending down of the AF-P phase boundary to be due to the critical fluctuations. Similar shiftings of phase boundaries are seen in spin-flop systems at bicritical points.[20] It was not possible to decide whether the λ line in FC2 is cut off by the first-order line or whether it meets the first-order line tangentially as at bicritical points. RG calculations of systems with CEP's do not show tangential behavior,[31] whereas Monte Carlo studies do not exclude such a behavior.[29] Figure 6 shows some examples of ω scans, the positions of which in the (H,T) plane are to be seen in Fig. 4. Because of the poor counting statistics at high fields we prefer to give a more qualitative interpretation of the profile analysis results. If we merely distinguish between continuous and discontinuous transitions this is no restriction. As long as we find Lorentzian-shaped contributions (points 1—3) there exist continuous transitions. Otherwise there is pure Bragg scattering at first-order transitions (point 4), and simple background scattering at the reciprocal-lattice position (010) (point 5) means the FI phase must exist there.

The situation for CC2 is more complicated; see Fig. 5. We find a triple point with coordinates $H_{TP}=3.972\pm0.009$ T and $T_{TP}=9.53\pm0.01$ K. Additionally, there exists another special point with coordinates $H_t=4.00\pm0.01$ T and $T_t=10.0\pm0.1$ K on the AF-P transition line. Let us call this point a tricritical one (TCP). Then we find $T_t-T_{TP}=0.47$ K, and this would be in agreement with MF theory where the calculations[13] give $T_t=\frac{2}{3}T_N$ and $T_t-T_{TP}=1.6$ K. Moreover, we found a very small overarch of the FI phase by the AF phase as in FC2. The temperature interval where this happens amounts only to 0.01 K instead of 0.3 K for FC2. Shifting of the AF-FI phase boundary by various experimental procedures, which we found and discussed for FC2, was not observed by us for CC2.

IV. EXPERIMENTS—CRITICAL BEHAVIOR

Although the critical scattering intensity in FC2 could be separated quite well from Bragg scattering, its overall contribution was very small. Therefore we could not get any reasonable exponents from it. CC2 showed no critical scattering at all. This is in accordance with measurements on $CoCl_2 \cdot 2D_2O$ (Ref. 33) which should be an even better candidate for observing critical scattering because of the missing incoherent hydrogen background scattering. Furthermore, for CC2, the small difference between the temperatures T_t and T_{TP} demonstrates that there was no chance to find exponents for the jumps in the sublattice magnetization and in the magnetization between the temperature and T_{TP}. Taking everything into account, we were restricted to the determination of the exponent β of the sublattice magnetization M_s. For this quantity, one has, at zero field, $M_s^2 \sim I_{Bragg} \sim \epsilon_T^{2\beta}$ $[\epsilon_T=(T_N-T)/T_N]$ and, in a field at constant temperatures, $M_s^2 \sim I_{Bragg} \sim \epsilon_H^{2\beta}$ $[\epsilon_H=(H_C-H)/H_C]$; H_C is the least-squares value of the transition-field strength of an isothermal field scan.

The critical exponent β of FC2 in zero field is found to be $\beta=0.30\pm0.01$ for $\epsilon_T<1.0\times10^{-1}$ as shown in Fig. 7. There also we can see the field dependence of M_s at a temperature $T=T_{CEP}+0.6$ K. Here, we found $\beta_{CEP}=0.29\pm0.02$ with $\epsilon_H<1.5\times10^{-2}$.

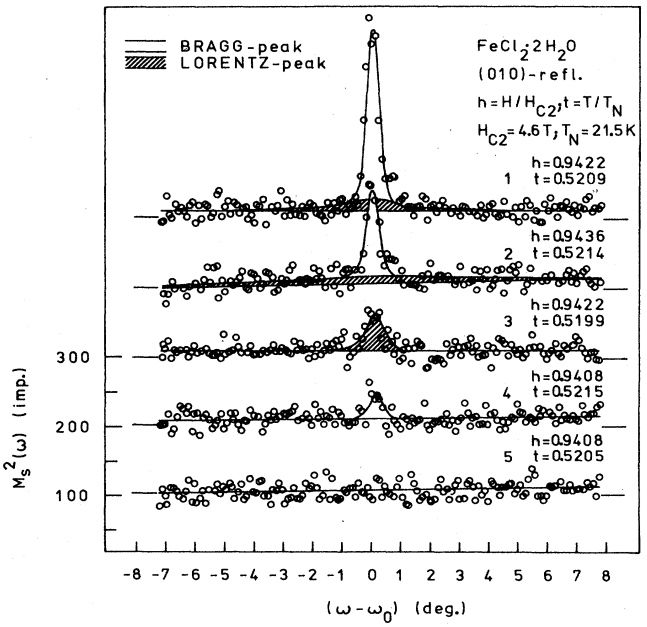

FIG. 6. ω scans of the reflection (010) of $FeCl_2 \cdot 2H_2O$ for the five different (H,T) coordinates as shown in Fig. 4; $\omega_0=0°$ for scan 1.

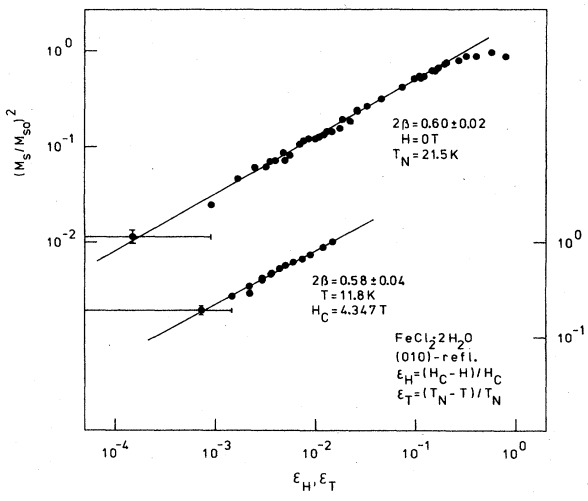

FIG. 7. Log-log plot of Bragg intensities of the reflection (010) of $FeCl_2 \cdot 2H_2O$ in zero field and for constant temperature $T=11.8$ K.

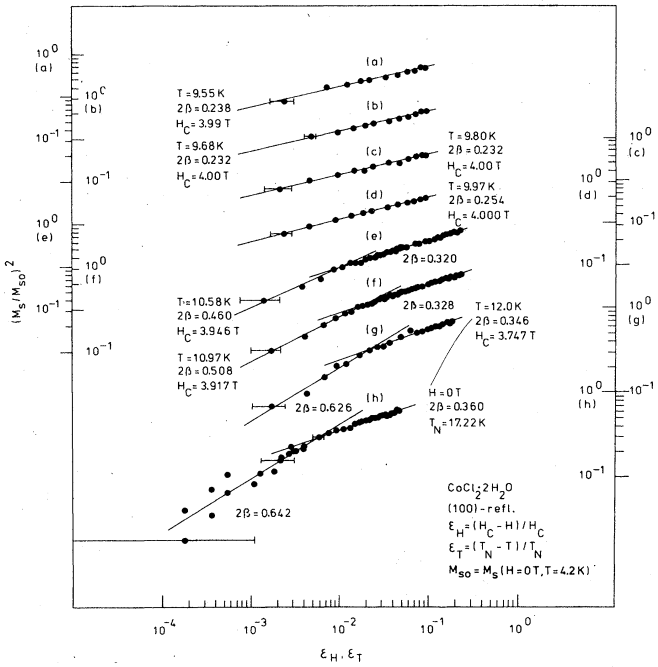

FIG. 8. Log-log plot of Bragg intensities of the reflection (100) of $CoCl_2 \cdot 2H_2O$ in zero field and for different constant temperatures.

Data to determine the critical exponents β for CC2 are presented in Fig. 8. The ϵ_T and ϵ_H ranges are shown therein and also in Fig. 5. At zero field near the Néel temperature ($\epsilon_T < 6\times10^{-3}$), we found $\beta_3 = 0.32\pm0.01$, whereas further away ($6\times10^{-3} < \epsilon_T < 6\times10^{-2}$) we found $\beta_1 = 0.18\pm0.01$. With increasing field, the critical region displaying β_3 becomes smaller and smaller until it disappears. This finally happens at the point (H_t, T_t), which we called TCP and where $\beta_t = 0.130\pm0.005$ is observed. This implies the existence of two crossover phenomena in this region, one from β_3 to β_t and one from β_1 to β_t, in addition to the crossover from β_3 to β_1 in zero field.

We want to emphasize that we did not transform our data by scaling methods such as the ones introduced by Fisher.[20] This is the reason for the apparent continuous course of the field dependence of the values of β_3 and β_1 with decreasing temperatures and increasing fields. Indeed, we cannot compare them directly with theory, but they clearly demonstrate the reliability of our experimental and mathematical evaluating methods. Near the CEP (H_{CEP}, T_{CEP}) for FC2 and at the point (H_t, T_t) for CC2, the AF-P lines in the diagrams are tangent to the horizontal which means $dH/dT = 0$. In this special case, an isothermal field scan in the (H,T) plane is perpendicular to the AF-P phase boundary, i.e., it represents a straight line coinciding with one scaling axis. Therefore, scaling is redundant, and we can compare directly our results for the exponents β_{CEP} and β_t with theoretical values, e.g., our measured β_t with $\beta_t = \frac{1}{4}$ for systems with $d \geq 3$. We suppose that the existence of points with $dH/dT = 0$ on the AF-P border is due to the intermediate FI phase. For other systems like $FeCl_2$ or $CsCoCl_3 \cdot 2D_2O$, without such an intermediate phase, scaling has to be carried out in any case for nonzero applied fields. Nevertheless, also in the case of $CsCoCl_3 \cdot 2D_2O$ the axis $[H - H_C(T)]/H_t$ was chosen as a "convenient choice of scaling fields."[7]

V. DISCUSSION—FC2

As mentioned above, FC2 shows neither tricritical nor bicritical behavior but a critical endpoint. In the literature there is, as far as we know, no reference to metamagnetic materials with similar critical behavior. The (p,T) diagram of He^4 exhibits a continuous λ line ending at two first-order transition lines in two separate so-called "upper and lower λ points."[32]

In the framework of mean-field theory, metamagnets with two exchange constants J and J' have been investigated theoretically.[27] The first one J is antiferromagnetic and couples the nearest neighbors; the second one J' is ferromagnetic and couples the next-nearest neighbors. Depending on the choice of the ratio J/J', there exist two different kinds of (H,T) phase diagrams, one with a tricritical point and a second one with a λ line ending on a first-order line. In particular, one can reproduce the principal topology of the λ line which we found for FC2. According to this theory, it ends on an AF-P first-order line at a so-called critical endpoint. As in FC2, but contrary to systems with tricritical points, the first-order line does not end at the CEP. In the case of the model, it goes into the AF phase ending therein at a so-called "bicritical endpoint"[27] (BCE) which might more properly be called a critical point (CP).[30,31] A third phase as in FC2 and, connected with it, a λ point or triple point do not exist in this theoretical phase diagram. As an experimental example for a system with a CEP, often $FeBr_2$ is nominated.[27] This is because the CEP is found in experiments[34] even though the BCE has not been found up to now. The experimentally determined exchange constants of $FeBr_2$ result in a ratio J/J' too far away from those which mean-field theory requires for the appearance of CEP's. In view of these last findings, the CEP of $FeBr_2$ is nowadays also regarded as a tricritical point.[35,36] It seems to us to be crucial that mean-field theory gives $\beta = \frac{1}{2}$ for the exponent at a critical point and at a critical endpoint, whereas the same theory yields $\beta_t = \frac{1}{4}$ at a tricritical point. We suggest measuring these exponents for $FeBr_2$.

RG theory of such a system with competing exchange interactions also finds the same exponents for critical points and critical endpoints;[31] that means no crossover over the whole λ line. All this together is similar to the situation we have found for FC2, i.e., a λ line ending on a first-order line without any crossover on this λ line. We also could not find a CP or BCE for FC2, perhaps because of the resolution, but perhaps because the FI phase displaces it.

There exists also a theoretical model with competing interactions in which mean-field theory gives a bicritical point.[29] By RG and Monte Carlo methods, it could be shown that fluctuations can drive the discontinuous transitions to temperatures beyond this bicritical point and thus change what was originally a bicritical point into a critical endpoint. This analysis also shows no crossover effects from zero-field exponents to other exponents on

the λ line including the CEP. This likewise brings us to the conclusion that we have observed for the first time a critical endpoint in a magnetic system.

VI. DISCUSSION—CC2

In zero field, we found $\beta_3=0.32$ for $\epsilon_T<6\times10^{-3}$ and $\beta_1=0.18$ for $6\times10^{-3}<\epsilon_T<6\times10^{-2}$. We can explain this result by a crossover in spatial dimension in zero field at $\epsilon_T=6\times10^{-3}$. This is suggested by the pronounced chain structure of the Co spins along the $\vec{c}$ axis. For comparison, we refer to former NMR experiments on CC2 which have yielded $\beta=0.30$ in zero field.[37] According to Fig. 3 of Ref. 37, this result is in fact restricted on two or three experimental data points on a straight line within $10^{-2}\leq\epsilon_T\leq2\times10^{-2}$. However, to get more data on this straight line one can fit these data with a lower value of T_N, e.g., $T_N=17.2$ K instead of $T_N=17.35$ K taken in this paper.[37] It results in an exponent β with $d<3$ behavior as we have observed it. The $d=3$ region which we have found for $\epsilon_T<10^{-3}$ has certainly not been detected in the NMR experiment, independently of whether one takes $T_N=17.35$ K or $T_N=17.2$ K.

At the TCP we found $\beta_t=0.13$. The exact value for $d\geq3$ is $\beta_t=\frac{1}{4}$ at a TCP. In the following we discuss three kinds of interpretation for our low experimental value β_t. First, as in zero field, there could be a crossover from $d=1$ behavior to the $d=3$ critical behavior also at the TCP. Second, one can suppose the effective dimensionality d is fixed at $d=3$ in the region of the TCP. In this case, one must in addition hypothesize the existence of impurities in the samples. Third, one can interpret the results by the existence of a critical endpoint instead of a TCP as we concluded for FC2. These three interpretations will be explained and then compared in the following.

First, in case of a crossover in spatial dimension, our experimental value of $\beta_1=0.18$ in zero field corresponds to an effective dimensionality of roughly $d_{\text{eff}}\simeq2.2$; i.e., $1<d<3$, because the exact $d=2$ value is $\beta_2=\frac{1}{8}$, and for $d=3$ the accepted value is $\beta_3=0.325$ (Table III of Ref. 38). Therefore our measured value β_1 does not correspond to true critical fluctuations, since because of universality arguments, only the exponents corresponding to dimensionalities $d=1$, 2, or 3 are significant. In any case, however, we can explain our measured value $\beta_1=0.18$ by an incompletely resolved crossover $d=3\rightarrow d=1$ at $\epsilon_T=6\times10^{-3}$. Below $\epsilon_T=6\times10^{-3}$, we find the usual $d=3$ behavior. With increasing fields, the region with $d=3$ behavior contracts, and the TCP is the point where we can observe it no longer. So, at the TCP, we can postulate a crossover to the "$d\simeq2.2$ state" together with a second crossover from standard critical behavior with $d\simeq2.2$ in zero fields to tricritical behavior with $d\simeq2.2$ at the TCP. Indeed it is known from $(3-\epsilon)$ expansions that the tricritical exponent β_t decreases with decreasing lattice dimensionality d $(d=3-\epsilon)$ according to[39]

$$\beta_t=\frac{1}{4}-\frac{\epsilon}{4}+\frac{3}{16}\frac{(n+2)(n+4)}{(3n+22)^2}\epsilon^2+O(\epsilon^3)\ .$$

Using this expansion together with a dimensionality $d_{\text{eff}}=2.5$, we would get the value for β_t we have found experimentally at the TCP. Certainly this expansion is not reliable for values of d that are too low; e.g., for $d=2$ it would give $\beta_t\simeq0$; however, Monte Carlo calculations[40] for a two-dimensional, antiferromagnetic Ising model yield $\beta_t\simeq0.06$. So, as a first and very rough estimate we can take from theory for the relation between the standard simple critical exponent β and the tricritical exponent β_t the expression $\beta=\beta_t+0.05$ for $2<d<3$. This is in accordance with our observed exponents $\beta=\beta_1=0.18$ and $\beta_t=0.13$.

The experimental values $\alpha_t=0.69$ and $\beta_t=0.126$ are known for the TCP of structural phase transitions of ND_4Cl (Ref. 41); furthermore, the values $\alpha_t=0.65$ and $\beta_t=0.15$ are found for the TCP of the metamagnet $CsCoCl_3\cdot2D_2O$.[7] This is in agreement with our value $\beta_t=0.13$. The exponent $\alpha_t=\frac{1}{2}+\frac{1}{2}\epsilon$ $(\epsilon=3-d)$ increases for $d<3$ beyond $\alpha_t=\frac{1}{2}$ or $d=3$.[39] This is also observed.[7,41] Therefore, for the last compound a lowered dimension $d<3$ was proposed.[10] Comparing the quasi-one-dimensional behavior of the magnetic specific heat of this compound[10] with that of CC2,[42] one can establish a temperature region with quasi-one-dimensional preorder also for CC2. This region lies only partially within the critical region but there it may influence the critical behavior both in zero field and at the AF-P phase boundary, but it will not result in exact $d=1$ critical exponents.

Some similar examples for a lattice-dimension crossover in zero field are known. As a $d=2\rightarrow d=3$ crossover it has been seen for Rb_2FeF_4 (Ref. 43) and K_2MnF_4 (Ref. 44). For K_2MnF_4, however, we must note that a second study[45] does not find the crossover and casts doubts upon the first investigation[44] because of impurities of the sample. Whether the crossover was detected or not, there must exist a crossover to $d=3$, since K_2MnF_4 shows ordinary $d=3$ behavior below T_N. The discussion for all these K_2NiF_4 compounds has been summarized as follows.[46] There exists a crossover temperature T^* which takes the value $0.97T_N$ for Rb_2FeF_4 and $0.996T_N$ for K_2MnF_4. For K_2NiF_4 itself, the crossover temperature T^* is so close to T_N that it was not possible to find $d=3$ behavior in experiments although it must exist. A lattice crossover from $d=2$ to 3 lying within the experimentally accessible part of the critical region was proved definitely for the compounds $(CH_3NH_3)_2CuCl_4$ and $(C_{10}H_{21}NH_3)_2CuCl_4$.[47]

There have been several discussions of the question of what kind of parameters determine whether the crossover temperature lies outside the critical region, within that part of the critical region which is accessible in experiments $(\epsilon_T>10^{-4})$, or too close to the critical point to be observable in experiments $(\epsilon_T<10^{-4})$. The discussion has been directed specifically towards CC2 as an example. According to the general treatments, a compound must fulfill some criteria to exhibit a lattice-dimension crossover within that part of the critical region which is experimentally accessible.[48] The crossover temperature is, for a given compound, not fixed; rather, this temperature depends on how it is measured. That means it differs for measurements of sublattice magnetization, susceptibility,

or magnetic specific heat. The crossover may not necessarily appear both below and above the critical point. The important factor is the ratio $R=J'/J$ of couplings J' of next neighbors between neighboring chains or planes to couplings J of next neighbors within chains or planes. Too large a value of R results in ordering with $d=3$ without a lower dimensional preordering; too small a value of R results in ordering with $d=1$ or 2 with the crossover to $d=3$ behavior happening too close to the critical point to be accessible in experiment. A necessary condition to observe crossover within the critical region is $J>0$.[48] This is realized for the Cu compound mentioned and for CC2 with its ferromagnetic spin chains, but not for K_2NiF_4. This is the reason that crossover to $d=3$ behavior could not be found in this compound.[43,48] Another analysis deals directly with CC2,[49] comparing experimental and calculated magnetic specific heat and susceptibility. The best agreement was found for $R=-0.2$, and a deviation from $d=3$ behavior in the direction of $d=1$ behavior could be seen clearly in experiment and theory. This was outside the critical region; the theory was not applied within the critical region. Further, Monte Carlo calculations on CC2 confirm these results.[50] Our experiments show that the theoretically and experimentally verified $d=1$ behavior extends into the critical region.

Second, besides the lowering of dimensionality, one has to discuss the so-called "random exchange" effects which are caused by impurities within the sample and influence the critical behavior. In zero field, there is crossover to new critical behavior,[51] but it develops so slowly that it usually is not observable. In applied fields, the situation changes drastically. A homogeneous external field which is applied to a uniaxial anisotropic antiferromagnetic system induces effective "random local fields" within the sample. These fields change critical behavior.[52] In systems with random fields and a spin dimensionality $n\geq 2$, RG theory yields deviations in critical behavior from mean-field theory for $d<6$ instead of the usual $d<4$. Long-range order disappears for $d<4$ instead of $d<2$.[53] For Ising systems ($n=1$) with random fields, long-range order is absent for $d<2$ instead of $d<1$ for pure systems. This is discussed in older[53] and recent works.[54,55] For tricritical behavior in random-field systems, RG theory yields deviations from mean-field theory for $d<5$ instead of $d<3$ as for pure systems.[56] Generally, one gets the tricritical exponents of d-dimensional random-field systems by taking the exponents of the corresponding pure system at $d-2$. This means that the ϵ expansion[39] for β_t is now valid for $\epsilon=5-d$ instead of $\epsilon=3-d$. We can take over our rough estimate for the dimensionality of our system CC2 and get now $d\simeq 2.5+2=4.5$. In other words, a system with fixed $d=3$ and random fields should result in a value for β_t much lower than our observed $\beta_t=0.13$. Therefore, we exclude random fields as significant for our sample of CC2.

Recently, earlier results for $FeCl_2$ (Refs. 4 and 5), which were obtained by neutron scattering, have been reinterpreted by introducing random fields.[57] This has been done to remove contradictions between these experiments[4,5] and optical investigations.[6] In principle, this work[57] is based on a critical dimension $d_1=3$ for breakdown of long-range order for Ising systems ($n=1$) with random fields. The assumption of $d_1=3$ is suggested by some theoretical papers[58–60] which consequently find the so-called $d\rightarrow d-2$ rule in the whole (n,d) plane. It follows that the random-field $d=3$ Ising model fails to show long-range order because the pure $d=1$ Ising model does not order.[58–60] The consequences for critical behavior in the (H,T) plane are enormous. In a random-field Ising system, there exists only a critical point T_N at zero field, but no λ line of continuous transitions between this critical point T_N and the point (H_t,T_t). The tricritical point (H_t,T_t) becomes a second, standard simple critical point.[57] With this theory and the additional assumption that the $FeCl_2$ sample in the neutron scattering experiments had contained impurities, but that the sample of the optical investigations did not, it was possible to find agreement between both experiments and between experiment and theory.[57] To us, this agreement relates only to temperatures below T_t. For temperatures above T_t and for fields $H>0$, such considerations would mean that long-range order with Bragg scattering could not be detectable. Neutron scattering, nevertheless, gives Bragg scattering, a line of continuous transitions between T_N and the tricritical point and even critical exponents β for $H>0$ (Ref. 5). Only if these last results are rejected can the assumption[57] of impurities in the $FeCl_2$ sample of neutron-diffraction work be accepted without contradiction. As these results have not been rejected, we believe that the existence of random fields in $FeCl_2$ is not demonstrated by experiments. Apart from this, there is the theoretical obscurity relative to the critical dimension $d_1=3$ or $d_1=2$ for random-field systems with $n=1$. Concerning CC2, we consider it decisive that we found Bragg scattering and exponents for temperatures $T_t<T<T_N$ in applied fields. In the framework of a critical dimension $d_1=3$ this could not be explained. Earlier results for $CsCoCl_3\cdot 2D_2O$ (Ref. 7) were also reinterpreted by introducing random fields.[57] Better agreement for temperatures below T_t results in strong contradictions for temperatures above T_t, because Bragg scattering was found in this region for this compound also.[7] Therefore, in our opinion, the observed deviation of the experimental tricritical exponent $\beta_t=0.13$ for CC2 from the theoretical value of $\beta_t=\frac{1}{4}$ cannot be explained by random-field effects. As shown above, it can be explained sufficiently by an effective spatial dimensionality $d<3$.

Third, we want to discuss the question of whether we can be sure that CC2 has a tricritical point together with a triple point or whether it has a λ line without a tricritical point but with a critical endpoint, until now denoted as a triple point. In the latter case, the exponent $\beta_t=0.13$ in nonzero field has only to be reinterpreted as a standard critical exponent $\beta_1=0.13$. Additionally we must assume that the smearing of the crossover which gives $\beta_1=0.18$ in zero field is reduced in a field so that the exponent β_1 approaches the real value for lower dimensionality in a field. Between the temperatures T_t of the tricritical point and T_{TP} of the triple point, we found a gap of only 0.47 K. In this region, in the case of the first interpretation, there would exist discontinuous transitions recognizable by the absence of critical scattering. As we could not detect crit-

ical scattering in the whole (H,T) diagram, we cannot use that fact to establish the nature of the transition. Furthermore, the temperature interval between T_t and T_{TP} is so small that jumps of the sublattice magnetization occur only within the critical region. In this region they would be detectable by bends in straight lines in log-log plots for small ϵ_H values. Such deviations from straight lines could also be caused by crossover from $d=1$ to 3, if the extension of the $d=3$ region in terms of the field coordinate is as small as we have found it. Therefore a crossover from $d<3$ to $d=3$ behavior is not distinguishable from a change from $d<3$ behavior to discontinuous transitions with jumps of the sublattice magnetization. Until now, the temperature T_t of the tricritical point was manifested by us as the lowest temperature for which we could detect a $d=3$ exponent. This happened in accordance with the definition of the tricritical point as that point at the end of a λ line where the high value of the simple critical exponent β changes to the low value of the tricritical exponent β_t. As discussed, we cannot exclude a very small $d=3$ region, not resolved by us, in the neighborhood of the AF-P phase boundary at a field of about 4.0 T and temperatures below T_t and above T_{TP}. In this case the point hitherto referred to as a tricritical point would coincide with the triple point, so in fact representing a critical endpoint. Its coordinates would be those hitherto attributed to the triple point, namely, $H_{CEP}=3.972 \pm 0.009$ T and $T_{CEP}=9.53 \pm 0.01$ K. We would interpret our experimental result as saying that, if a tricritical point exists in CC2, it is situated at a temperature 0.47 K above the triple point at most. It is still possible that the tricritical point does not exist and the triple point of CC2 is in fact a critical endpoint as found for FC2. As discussed, one cannot distinguish the two cases by the measured value of β_t. This must be studied in further experiments.

VII. CONCLUSIONS

To summarize the experiments and the discussion up to this point, we have established for FC2 a λ line ending on the FI phase boundary in a critical endpoint. In the case of CC2 we would interpret our experiments in the same way but we could not totally exclude the existence of a tricritical point on the AF-P phase boundary dividing this line in two parts, a λ line going to the Néel point and a first-order line meeting the FI phase boundary in a triple point. The reason for our difficulties comes from a crossover in spatial dimension, which we found in zero field and which we could follow nearly up to the FI phase. This crossover in zero field is predicted by theoretical treatments in the literature for the case of our compound CC2. Nothing is known about a similar crossover in FC2. Our experiments indeed show that this effect is absent for FC2. The question arises as to whether this can be understood in terms of differences in the values of the exchange constants or even in the Hamiltonians for the two compounds. Generally, and in brief, it is true that the Co compound must be described by a strong anisotropy of exchange constants whereas the Fe compound can be described by isotropic exchange constants together with a single-ion term D.[61-63] As one example of the effect of these differences, the anisotropies make the $\vec{b}$ direction an easy axis for CC2 and the crystal-field effects make the $\vec{a}$ direction an easy axis for FC2. As a second example, the anisotropies cause the well-known spin-cluster resonances which are found only for CC2.[63] In the present work, we could observe differences in the critical behavior coming from these different Hamiltonians.

Incidentally, it is known that such a single-ion or D term does not change the critical behavior, provided it is not too strong.[64] If it is strong, it is not known in detail what happens. For that case for ferromagnets only, it was shown that the D term can drive the system to a new zero-field tricritical point if certain assumptions are made about the ratio of the exchange constants to the D term.[65] We could not detect any unusual critical behavior in zero field in our experiment on FC2. It would be desirable to study theoretically a system such as FC2 with competing exchange interactions together with a single-ion term. With respect to our question for modern theories at the end of Sec. I, we want to point to recent Monte Carlo calculations of a phase diagram of an antiferromagnetic Ising system with two competing exchange interactions of different sign, in which for the first time the FI phase could be reproduced by a theory including critical phenomena.[66]

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[1]R. B. Griffiths, Phys. Rev. Lett. **24**, 715 (1970).
[2]E. K. Riedel and F. J. Wegner, Phys. Rev. Lett. **29**, 349 (1972).
[3]F. J. Wegner and E. K. Riedel, Phys. Rev. B **7**, 248 (1973).
[4]R. J. Birgeneau, G. Shirane, M. Blume, and W. C. Koehler, Phys. Rev. Lett. **33**, 1098 (1974).
[5]R. J. Birgeneau, in *Magnetism and Magnetic Materials—1974 (San Francisco), Proceedings of the 20th Annual Conference on Magnetism and Magnetic Materials,* edited by C. D. Graham, G. H. Lander, and J. J. Rhyne (AIP, New York, 1975), p. 258.
[6]J. F. Dillon, E. Y. Chen, and H. J. Guggenheim, Phys. Rev. B **18**, 377 (1978).
[7]A. L. M. Bongaarts and W. J. M. de Jonge, Phys. Rev. B **15**, 3424 (1977).
[8]E. Stryjewski and N. Giordano, Adv. Phys. **26**, 487 (1977).
[9]A. van der Bilt and A. J. van Duyneveldt, Physica (Amsterdam) **95B**, 305 (1978).
[10]K. Kopinga, Q. A. G. van Vlimmeren, A. L. M. Bongaarts, and W. J. M. de Jonge, Physica (Amsterdam) **86-88B**, 671 (1977).

[11]Q. A. G. van Vlimmeren, C. H. W. Swüste, W. J. M. de Jonge, M. J. H. van der Steeg, J. H. M. Stoelinga, and P. Wyder, Phys. Rev. B **21**, 3005 (1980).
[12]H. Weitzel and W. Schneider, Solid State Commun. **14**, 1025 (1974).
[13]K. Yamada and J. Kanamori, Progr. Theor. Phys. **38**, 541 (1967).
[14]W. Schneider and H. Weitzel, Solid State Commun. **13**, 303 (1973).
[15]W. Schneider and H. Weitzel, Acta Crystallogr. Sect. A **32**, 32 (1976).
[16]W. Schneider and H. Weitzel, Solid State Commun. **18**, 995 (1976).
[17]A. Narath, Phys. Lett. **13**, 12 (1964).
[18]A. Narath, Phys. Rev. **139**, A1221 (1965).
[19]I. S. Jacobs and P. E. Lawrence, Phys. Rev. **164**, 866 (1967).
[20]M. E. Fisher, in *Magnetism and Magnetic Materials—1974 (San Francisco), Proceedings of the 20th Annual Conference on Magnetism and Magnetic Materials,* edited by C. D. Graham, G. H. Lander, and J. J. Rhyne (AIP, New York, 1975), p. 273.
[21]L. Graf, Thesis, Technische Hochschule Darmstadt, 1977.
[22]H. Langhof, H. Weitzel, E. Wölfel, and W. Scharf, Acta Crystallogr. Sect. A **36**, 741 (1980).
[23]K. Katsumata, J. Phys. Soc. Jpn. **39**, 42 (1975).
[24]K. Katsumata, Physica (Amsterdam) **86-88B**, 1124 (1977).
[25]L. Graf, J. Magn. Magn. Mater. **6**, 124 (1977).
[26]A. F. G. Wyatt, J. Phys. C **1**, 684 (1968).
[27]J. M. Kincaid and E. G. D. Cohen, Phys. Lett. **50A**, 317 (1974).
[28]M. Kerszberg and D. Mukamel, Phys. Rev. Lett. **43**, 293 (1979).
[29]J. R. Banavar, D. Jasnow, and D. P. Landau, Phys. Rev. B **20**, 3820 (1979).
[30]S. Galam and A. Aharony, J. Phys. C **13**, 1065 (1980).
[31]T. A. L. Ziman, D. J. Amit, G. Grinstein, and C. Jayaprakash, Phys. Rev. B **25**, **319** (1982).
[32]H. A. Kierstead, Phys. Rev. **162**, 153 (1967).
[33]J. K. Kjems, J. Als-Nielsen, and H. Fogedby, Phys. Rev. B **12**, 5190 (1975).
[34]C. Vettier, H. L. Alberts, and D. Bloch, Phys. Rev. Lett. **31**, 1414 (1973).
[35]W. B. Yelon and C. Vettier, J. Phys. C **8**, 2760 (1975).
[36]D. Blankschtein and A. Aharony, Phys. Rev. B **26**, 415 (1982).
[37]H. Yamakawa, J. Phys. Soc. Jpn. **47**, 763 (1979).
[38]J. C. le Guillou and J. Zinn-Justin, Phys. Rev. B **21**, 3976 (1980).
[39]M. J. Stephen and J. L. McCauley, Phys. Lett. **A44**, 89 (1973).
[40]D. P. Landau, J. Tombrello, and R. H. Swendsen, in *Proceedings of the International Conference on Ordering in Two Dimensions, Lake Geneva, 1980,* edited by S. K. Sinha (Elsevier North-Holland, Amsterdam, 1980), p. 351.
[41]C. W. Garland, D. E. Bruins, and T. J. Greytak, Phys. Rev. B **12**, 2759 (1975).
[42]T. Shinoda, H. Chihara, and S. Seki, J. Phys. Soc. Jpn. **19**, 1637 (1964).
[43]R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B **1**, 2211 (1970); see also Ref. 46, in particular Fig. 48, p. 129.
[44]H. Ikeda and K. Hirakawa, J. Phys. Soc. Jpn. **33**, 393 (1972).
[45]R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B **8**, 304 (1973).
[46]L. J. de Jongh and A. R. Miedema, Adv. Phys. **23**, 1 (1974).
[47]L. J. de Jongh, Physica (Amsterdam) **82B**, 247 (1976).
[48]L. J. de Jongh and H. E. Stanley, Phys. Rev. Lett. **36**, 817 (1976).
[49]R. Navarro and L. J. de Jongh, Physica (Amsterdam) **94B**, 67 (1978).
[50]T. Graim and D. P. Landau, Phys. Rev. B **24**, 5156 (1981).
[51]C. Jayaprakash and H. J. Katz, Phys. Rev. B **16**, 3987 (1977).
[52]S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979).
[53]Y. Imry and S. Ma, Phys. Rev. Lett. **35**, 1399 (1975).
[54]G. Grinstein and S. Ma, Phys. Rev. Lett. **49**, 685 (1982).
[55]J. Villain, J. Phys. Lett. **43**, L551 (1982).
[56]A. Aharony, Phys. Rev. B **18**, 3318 (1978).
[57]R. J. Birgeneau and A. N. Berker, Phys. Rev. B **26**, 3751 (1982).
[58]E. Pytte, Y. Imry, and D. Mukamel, Phys. Rev. Lett. **46**, 1173 (1981).
[59]K. Binder, Y. Imry, and E. Pytte, Phys. Rev. B **24**, 6736 (1981).
[60]D. Mukamel and E. Pytte, Phys. Rev. B **25**, 4779 (1982).
[61]K. A. Hay and J. B. Torrance, Phys. Rev. B **2**, 746 (1970).
[62]L. Graf, Phys. Status Solidi (b) **88**, 429 (1978).
[63]J. B. Torrance and M. Tinkham, Phys. Rev. **187**, 595 (1969).
[64]A. Aharony, *Magnetism and Magnetic Materials—1973 (Boston), Proceedings of the 19th Annual Conference on Magnetism and Magnetic Materials,* edited by C. D. Graham and J. J. Rhyne (AIP, New York, 1974), p. 863.
[65]D. M. Saul, M. Wortis, and D. Stauffer, Phys. Rev. B **9**, 4964 (1974).
[66]K. Binder, W. Kinzel, and W. Selke, J. Magn. Magn. Mater. **31-34**, 1445 (1983).