Phase diagram (T, H) investigation by direct measurement of dR/dT in a magnetic material

Ph. Gandit, F. Terki, J. Chaussy, P. Lejay

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Phase diagram (T, H) investigation by direct measurement of dR/dT in a magnetic material

Ph. Gandit, F. Terki, J. Chaussy and P. Lejay

Centre de Recherches sur les Très Basses Températures, 25 avenue des martyrs, BP 166, 38042 Grenoble Cedex 09, France

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Résumé. — La dépendance en température de la résistance $R(T)$ et de la partie singulière de $\frac{\mathrm{d}R}{\mathrm{d}T}$ au voisinage des régions critiques, sous champs magnétiques compris entre 0 et 5 T sont présentées dans cet article. Ces mesures ont été obtenues à l'aide d'une nouvelle technique qui permet d'accéder directement à $\left.\frac{\mathrm{d}R(T)}{\mathrm{d}T}\right|_H$ Cette quantité qui est reliée au coefficient de transport $\frac{1}{R}\,\frac{\mathrm{d}R}{\mathrm{d}T}$, est proportionnelle à la chaleur spécifique au voisinage des transitions de phases. La précision des données expérimentales nous a permis de vérifier le bon accord entre l'analyse que nous avons menée pour $H = 0$ T, et la valeur des exposants critiques prédite par différents modèles de la théorie du groupe de renormalisation. En outre, nous présentons une nouvelle approche de la détermination d'un diagrame de phase (T, H) en utilisant les propriétés de transport $R(T,\ H)$ et $\left.\frac{\mathrm{d}R(T)}{\mathrm{d}T}\right|_H$ d'un monocristal d'erbium, au voisinage des températures critiques.

Abstract. — The temperature dependences of the resistance $R(T)$ and the singular part of $\frac{\mathrm{d}R}{\mathrm{d}T}$ near the critical regime and in the presence of magnetic fields H [0-5 T] are presented in this paper. The measurements were carried out using a novel technique from which $\left.\frac{\mathrm{d}R}{\mathrm{d}T}\right|_H$ can be determined directly. This quantity, being related to the transport coefficient $\frac{1}{R}\,\frac{\mathrm{d}R}{\mathrm{d}T}$, is directly connected to the specific heat behavior near the phase transition. The high accuracy of our experimental data allows us to compare the critical exponents deduced from our previous measurements in zero field ($H = 0$ T) and those predicted by several models of the renormalization group theory. We present a complete phase diagram for a single crystal of erbium oriented along the c-axis. This constitutes the first systematic study of field-induced magnetic phase transitions using the magnetoresistance $R(T,\ H)$ and its derivative $\left.\frac{\mathrm{d}R}{\mathrm{d}T}\right|_H$ near the critical points.

Introduction.

The effect of critical fluctuations on the electrical resistance of magnetic materials became a subject of widespread interest several years ago. Previous work has shown that the temperature dependence of the resistance in the vicinity of the critical temperature T_N is directly related to the magnetic energy [1-3]. Suezaki and Mori [4] studied the role of critical fluctuations in antiferromagnets and concluded that long range correlations should dominate approaching the paramagnetic state, from which $\frac{\mathrm{d}R(T)}{\mathrm{d}T} \propto t^{2\beta-1} (t = |1 - T/T_N|)$. Then, Fisher [2, 5] and Langer [2] predicted that the magnetic contribution to the temperature derivative of the electrical resistance near the Curie temperature $\mathrm{d}R(T)/\mathrm{d}T$, should be proportional to the specific heat, $C(T)$, for a ferromagnet, since the temperature dependence of both quantities is dominated by short-range part of the spin-correlation function. This theoretical prediction has been quantitavely verified in the quasi-itinerant ferromagnets nickel [6] and iron [8], and in β-brass [9], an order-disorder system.

However, the situation is much more uncertain for the case of antiferromagnets. Although, the conclusion that $\frac{\mathrm{d}R(T)}{\mathrm{d}T} \propto C(T)$ has been extended to antiferromagnets below the Néel temperature $T \leq T_{\mathrm{N}}$ [6, 10, 11]. Then, Richard and Geldart [11], and Alexander *et al.* [12] have shown that for $T > T_{\mathrm{N}}$, long range correlations play a role (prediction based on the Ornstein-Zernicke approximation for spin correlations $\frac{\mathrm{d}R(T)}{\mathrm{d}T} = t^{1/2}$) [12]. They also discussed in some detail the effect of critical fluctuations on the electrical resistivity of antiferromagnets and binary alloys. In recent work [13], we presented a numerical analysis of the temperature derivative of the electrical resistance along the c-axis of a single crystal of Er in zero field. The high accuracy of our $\frac{\mathrm{d}R}{\mathrm{d}T}$ measurements enabled us to compare our experimental results with theoretical predictions. Our analysis is based on the physical expectation that the resistance and its derivative with respect to temperature are continuous at T_{N}.

Moreover, we found that the physical requirement such as the universality of the critical amplitude ratio $\frac{A}{A'}$, given by recent group renormalization results [14], should be fulfilled. This more sophisticated analysis of the experimental data near the critical regime had been suggested by Balberg and Maman [15], and Malmström and Geldart [16]. They confirmed that a different analysis of the critical resistance behavior yields different critical exponents for similar antiferromagnet systems [15-20]. Moreover, we have suggested [13] previously that far from the critical regime, $\frac{\mathrm{d}R}{\mathrm{d}T}$ follows an exponential law, which takes into account the excitations of the spin system. In the present paper we focus on the magnetic as well as the temperature dependence of the critical resistance $R(T, H)$. We present high precision data for $\left.\frac{\mathrm{d}R(T)}{\mathrm{d}T}\right|_H$ measured by a new technique which we have developed to directly determine this transport property [21]. In the literature, the magnetoresistance of nickel near its Curie temperature has to date been considered only in the mean field approximation by Schwerer [22]. Simon and Salomon [8] attempted to measure both the specific heat and the resistivity of gadolinium near the Curie point as function of magnetic field and show that they were proportional. Recent developments in the understanding of the critical behavior of the resistance in magnetic materials for zero field and under applyied magnetic fields have been reviewed [23-25]. In this paper we extend our previous work [13] to find fields induced magnetic phase transitions in an antiferromagnetic single crystal of erbium.

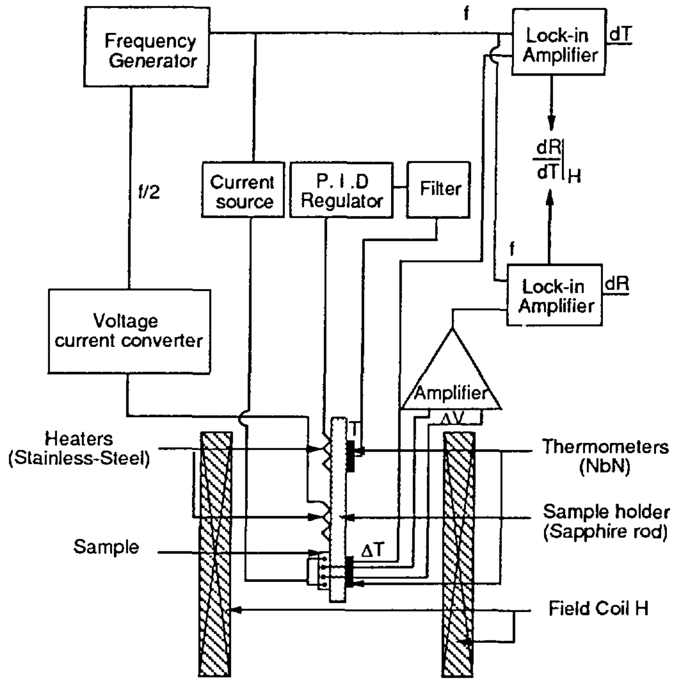

Fig. 1. — Schematic diagram of the experimental set-up.

Measuring principle [13, 21].

The experimental method consists of measuring the response of the sample resistance $R_s(T)$ to a small temperature oscillation ΔT_{ac}, from which the derivative of the resistance with respect to temperature $\frac{\mathrm{d}R_s(T)}{\mathrm{d}T}$ is found. The sample we want to study is placed in the center of a superconducting field coil, which enabled us to determine the features of the magnetoresistance of the sample, up to a field $H = 5$ T. The sample is biased by a DC current I_{dc} at a regulated temperature T. The DC voltage V_{dc} which appears at its extremities, is proportional to its resistance $R_s(T)$. Then, an oscillation of the temperature ΔT_{ac}, induces an additional AC voltage ΔV_{ac} (proportional to the variation of the resistance $\Delta R_s(T)$), given by:

$$V_{dc} + \Delta V_{ac} = R_s(T,\ H)\ I_{dc} + \Delta R_s(T,\ H)\ I_{dc} \tag{1}$$

We can then deduce the quantitv $\left.\frac{\mathrm{d}R_s(T)}{\mathrm{d}T}\right|_H$

$$\left.\frac{\mathrm{d}R_s(T)}{\mathrm{d}T}\right|_H = \left.\frac{\Delta R_s(T)}{\Delta T_{ac}}\right|_H = \frac{1}{I_{dc}}\frac{\Delta V_{ac}}{\Delta T_{ac}} \tag{2}$$

Apparatus [21].

A schematic diagram of the experimental set up is given in figure 1.

- The average temperature of the system (sample and sample holder) is regulated by a PID (proportional, integral and derivative) controller associated with a heater-thermometer pair.
- The temperature modulation ΔT_{ac} is imposed using a second resistance heater driven by a voltage to current converter at a frequency $f/2 \sim 2$ Hz.
- The second thermometer is biased by a direct current. Thus the DC voltage which appears across this thermometer allows us to determine accurately the sample's average temperature. At the same time, an alternating voltage component also appears, it enables the temperature oscillation ΔT_{ac} to be measured.
- NbN thermometers were chosen because of their reproducibility and sensitivity over the temperature range of interest. Moreover, we have measured the magnetic field dependence of the NbN resistance and checked that the deduced temperature T is only slightly modified by applying a magnetic field ($\Delta T/T < 2$ % at $T \sim 4.2$ K for a field of 5 T).
- The two AC voltages, which correspond to the temperature oscillation ΔT_{ac} and the variation of the resistance $\Delta R_{\mathrm{s}}(T)$ respectively are detected after amplification by two lock-in amplifiers operating at the same frequency $f (f = 4$ Hz).

A more complete description of the measurement technique can be found in reference [21]. In addition to direct physical interpretation [13] of the measurements, we would like to emphasize again the high accuracy of the results obtained by our technique. Indeed a resolution $\frac{\Delta R}{R}$ of $10^{-8}(\mathrm{Hz})^{-1/2}$ with R equal to a few mΩ can be achieved. By averaging over 100 s this resolution increases to 10^{-9} which is much more accurate than a direct measurement of the resistance.

Experimental results and physical interpretation.

The sample was a single crystal of erbium grown by the Czochralski method under a purified atmosphere of argon. The resistance and temperature resistance derivative data were measured parallel to the c-axis, with magnetic fields applied along the easy axis of magnetization (c-axis).

1. MEASUREMENTS OF $R(T)$ AND $\mathrm{d}R/\mathrm{d}T$ WITHOUT APPLYING A MAGNETIC FIELD [13]. — Recent measurements of the resistance as a function of the temperature within the range [10 K – 300 K] confirmed that Erbium exhibits three distinct types of spin ordering. We have studied the behavior of the resistance of a single crystal of Er for ($H = 0$ T) within the temperature interval [10 K – 300 K]. A sharp increase in the electrical resistance with decreasing temperature occurs at the Néel temperature, $T_{\mathrm{N}} = 87.6$ K, and characterizes the para-antiferromagnetic transition. Then the resistance increases with decreasing temperature down to $T = 60$ K where it reaches its maximum value. Between the Néel temperature and this intermediate temperature, $T_{\mathrm{H}} = 60$ K, the magnetic structure is "modulated along the c-axis". Below T_{H} and down to the Curie temperature of 20 K, the resistance drops quickly, due to another type of antiferromagnetic rearrangement, with a complex structure [26, 27]. Below T_{c}, the magnetic moments order in a ferro-conical structure [28]. We also noted a distinct hysteresis in the resistance curve in the temperature range [10 K – 60 K] which disappeared above 60 K. Figure 2 shows our temperature derivative measurements $\frac{\mathrm{d}R}{\mathrm{d}T}$ (points), since the solid lines are fits for different temperature range. The physical interpretation of such laws are discussed in detail in reference [13].

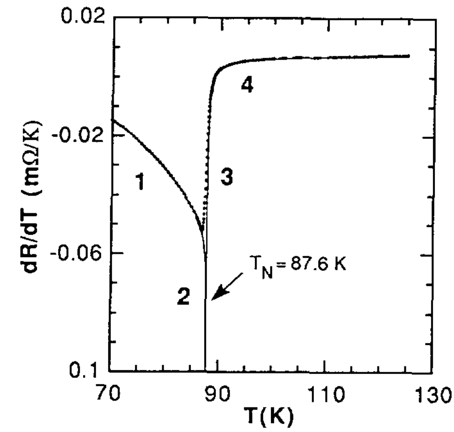

Fig. 2. — Temperature derivative of the resistance plotted *versus* temperature for Er. Solid lines are fits for different temperature ranges as follows : 1) $\frac{dR}{dT} = AT^2\, e^{-\Delta/T}$ with $\Delta \approx 3T_N$ for $T \ll T_N$, 2) $\frac{dR}{dT} = B' + A'(1 - T/T_N)^{-\alpha'}$ for $T < T_N$ in the close vicinity of T_N, with $\alpha' = 0.001$, 3) $\frac{dR}{dT} = B + A\,(T/T_N - 1)^{-\alpha}\left(1 + F\,(T/T_N - 1)^{\Delta_1}\right)$ for $T > T_N$ in the close vicinity of T_N, with $\alpha = 0.001$, $\Delta_1 = 0.55$ and, 4) $\frac{dR}{dT} = B + A\,(T/T_N - 1)^{-\alpha}$ for $T \gg T_N$ with $\alpha = 0.5$ (mean field exponent).

2. MEASUREMENTS OF $R(T)$ IN THE PRESENCE OF MAGNETIC FIELDS. — Figure 3 shows resistance measurements in the presence of magnetic fields, within the range [0 – 3 T], as the temperature is increased from 10 K to 300 K. The bump of the resistance observed at $T = 60$ K in zero field decreases continuously when the magnetic field is increased, and has practically disappeared in a magnetic field of 2.6 T. This effect has already been observed in a similar antiferromagnet (Tb) [29]. In order to scrutinize this feature we measured the thermal dependence of the c-axis resistance for various values of the magnetic field // c in the range [2 T – 3 T] (Fig. 4). We then deduced the value of the field ($H \sim 2.6$ T) for which the maximum of the resistance vanishes. We also noted that the hysteresis observed for zero field in the temperature range [10 K-60 K] tends to disappear for a field of about 2.5 T. Figure 5 shows the effect of different values of magnetic fields 1, 1.5, 2 and 2.5 T on the hysteretic behavior of the resistance within the temperature interval [10-60K]. For higher fields in the range [3-5 T] (Fig. 6), the antiferromagnetic phase is destroyed while at the same time a marked change of slope occurs as the field is increased (Fig. 6). This feature can be associated with the transition from the paramagnetic to the ferromagnetic phase. Indeed, recent neutron maesurements in the presence of magnetic fields [30] confirm that no antiferromagnetic phase persists for fields above 2.5 T.

3. MEASUREMENTS OF dR/dT IN PRESENCE OF MAGNETIC FIELDS. — Figure 7 shows our $\frac{dR}{dT}$ measurements in applied magnetic fields along the axis of easy magnetization (c-axis) in the temperature range 70 K - 100 K. The paramagnetic-antiferromagnetic transition which occurs at the Néel temperature (87.6 K in zero field) moves towards lower temperatures with increasing magnetic field from 1 T to 2 T. Moreover, for a field of 2 T a second inflexion point begins to appear in the vicinity of the critical regime. Then, at a field of 2.25 T, a splitting

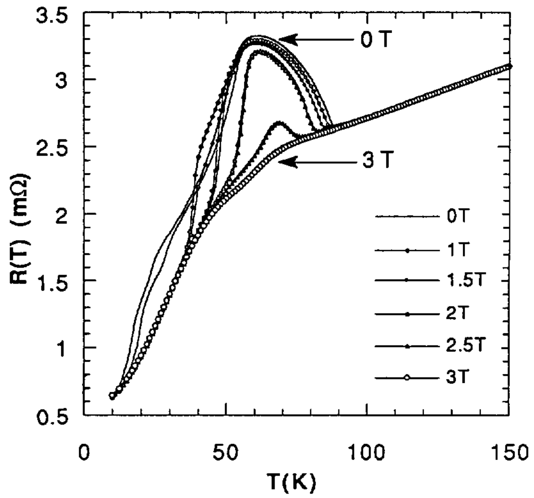

Fig. 3. — c-axis resistance of single crystal of Er *versus* temperature for different magnetic fields applied along the c-axis ($0 < H < 3$ T).

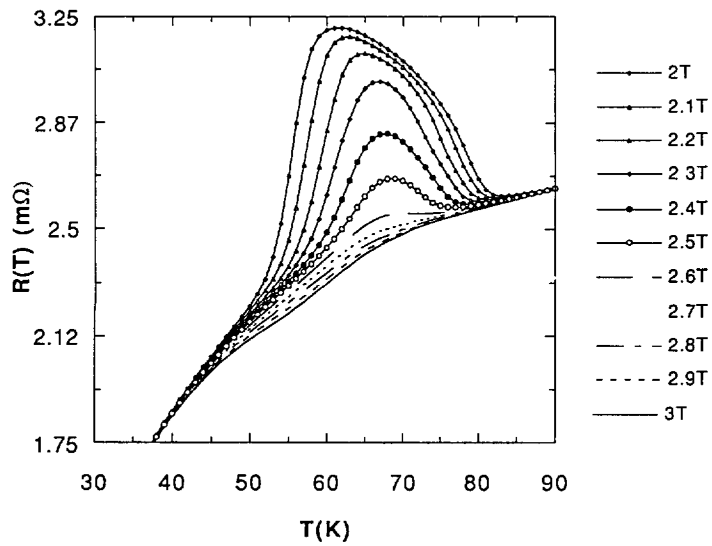

Fig. 4. — Detailed measurements of the magnetoresistance of Er for fields ranging from 2 T to 3 T.

of the critical temperature is clearly observed. This feature indicates that a new phase with an another type of order appears. This constitutes the first systematic evidence, by transport property measurements $\frac{\mathrm{d}R}{\mathrm{d}T}$, of a new magnetic rearrangement induced by an external magnetic field. The physical interpretation of this experimental singularity is that a field of 2 T tends to order the magnetic moments parallel to the direction of the field in the antiferromagnetic phase below T_{N}. Thus the transition from the paramagnetic state to the antiferromagnetic

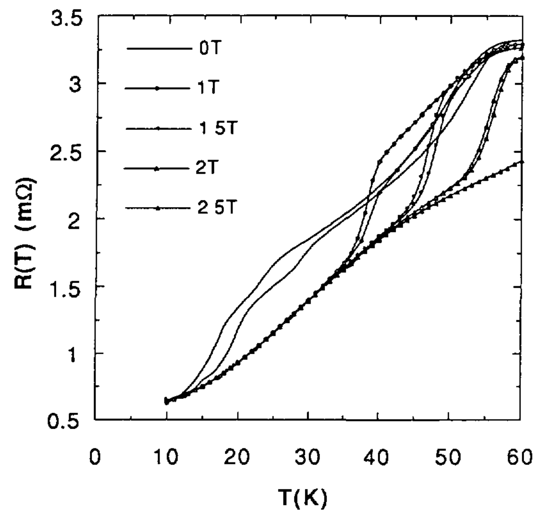

Fig. 5. — Hysteresis behavior of the resistance in temperature range [10 - 60 K] in magnetic fields ($0 < H < 2.5$ T).

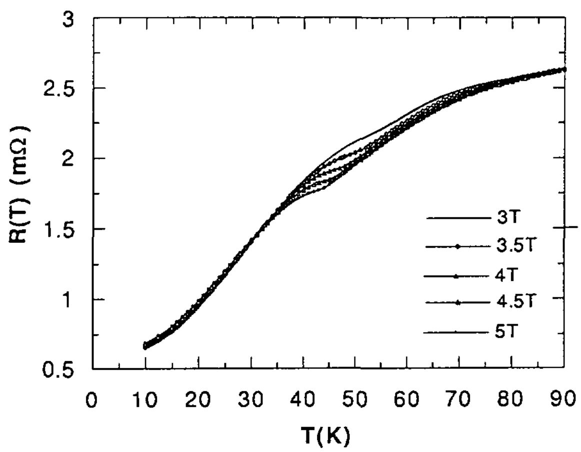

Fig. 6. — Resistance *versus* temperature curves for high fields ($3 < H < 5$ T).

(c-axis modulated) ordering proceeds *via* an intermediate ferromagnetic ordering as confirmed by neutron studies [30].

The second anomaly occuring just below the above corresponds to the transition from ferromagnetic (c-axis modulated structure?) to the modulated antiferromagnetic phase. Even so, a critical field of about 2.45 T is sufficient to completely destroy the c-axis modulated antiferromagnetic structure. Indeed, the corresponding anomaly is progressively attenuated and disappears at about this field (Fig. 7). Nevertheless the singularity which still persists at fields greater than this critical field characterizes the transition from the paramagnetic state to the new ferromagnetic ordering discussed above. This field also corresponds to the disappearance

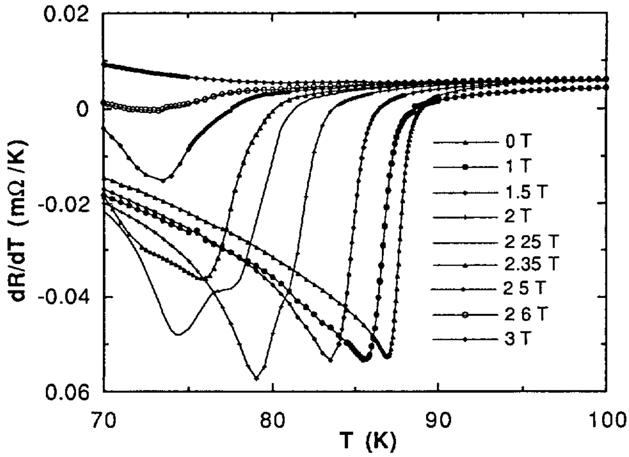

Fig. 7. — Temperature derivative of the resistance plotted against temperature between 70 and 100 K for different magnetic fields ($0 < H < 3$ T) showing the para-antiferromagnetic transition. Note the splitting of the peak at 2.25 T.

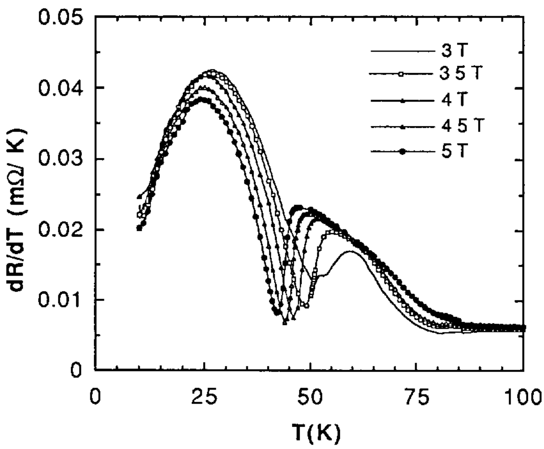

Fig. 8. — $\frac{dR}{dT}$ measurements *versus* temperature for high magnetic fields ($3 < H < 5$ T) showing the para-conical ferromagnetic transition.

of the bump in the resistance behavior. This last transition from the helical antiferromagnetic phase to the conical ferromagnetic order moves towards higher temperatures under applied fields of up to 2.45 T. Although, for fields greater than 2.6 T, the phase boundary between the paramagnetic state and the conical ferromagnetic structure is displaced towards lower temperatures. Figure 8 shows our $\frac{dR}{dT}$ measurements in the presence of magnetic fields ranging from 3 T to 5 T. The Curie temperature has been taken at the first inflexion point between the two peaks, decreasing the temperature, on the $\frac{dR}{dT}$ curves under different magnetic fields.

As this work progressed, a paper on the magnetic structured of Er deduced by neutron scattering in applied magnetic fields [30] confirmed our prediction of the phase diagram (Fig. 9). We emphasize that our study extends to 5 T and therefore includes the para-conical ferromagnetic phase boundary. Moreover the new ferromagnetic domain is defined on one side by the para-ferro line and on the other side by the ferro-antiferro modulated phase (Fig. 9). These lines have not been clearly defined in the very recent neutron studies [30] or by ultrasound measurements of Er [31].

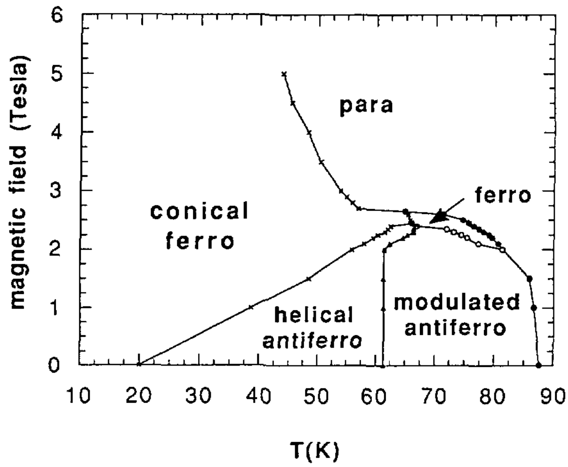

Fig. 9. — Schematic $(H-T)$ phase diagram for erbium deduced from $R_{\rm H}$ and $\frac{{\rm d}R}{{\rm d}T}\big|_H$ curves.

Conclusion.

The high sensitivity of the technique we have developed enabled us to detect a small change in the spin ordering under magnetic field in the range [2 - 2.6 T]. That this structure had not been predicted *a priori* constitutes irrefutable evidence of the utility of measurement of $\frac{{\rm d}R}{{\rm d}T}$. Another study of the magnetization of Er [32] gave an experimental value of the critical field of 2.24 T for which the ferro-paramagnetic phase transition occurs. Whilst, Gama and Foglio [32] also mentioned that at 65 K the model proposed by Jensen [33] predicts ferromagnetic order at a field of 2.6 T, although these deduced critical fields (2.24 and 2.6 T) are associated with two different ferro-paramagnetic phase transitions. The first transition which has been observed by the magnetization measurements at a field of 2.24 T is also marked on $\frac{{\rm d}R}{{\rm d}T}$ measurements and can be attributed to the splitting of the critical temperature and may characterize the transition between the paramagnetic state to the new ferromagnetic ordering (Fig. 9). However, the second critical field of 2.6 T at $T = 65$ K predicted by Jensen exactly constitutes the beginning of the para-ferromagnetic (conical structure) line transition in our phase diagram. Moreover, we emphasize that our previous theoretical analyses for $H = 0$ T [13] has suggested an activated law over a large temperature range below and far from $T_{\rm N}$, which indicates a possible existence of an energy gap. This gap may be attributed to the effects of inelastic scattering from magnons and phonons. To extend this work, detailed theoretical analyses of the recent measurements under applied magnetic fields will be presented in a forthcoming publication [34]. This will confirm the predicted power-laws for the temperature dependence of the magnetoresistance of antiferromagnetic metals [25] within the critical regime and the mean-field regime, and our suggested activated law below and far from T_N.

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