hf field is caused by relaxation effects.[28] This is very unlikely. Down to about 0.4 of the saturation field, we observe well-resolved spectra corresponding to a unique hf field. This means that the Eu-ion relaxation rate is fast enough to average over the various exchange-split ground-state energy levels. If the relaxation slowed down severely near T_t, this would show up in a broadening of the lines and in Mössbauer-absorption patterns consisting of complex broad spectra. Since we see no evidence of this, we can rule out the possibility of relaxation effects contributing to the shape of the observed spectra around T_t.

We can use the Clausius-Clapeyron equation in the form $dT_t/dP=\Delta V/\Delta S$ to relate the volume change determined from the thermal-expansion results to the entropy change and pressure dependence of the first-order transition temperature. Using our value of $\Delta V/V=2.1\times10^{-4}$ and Gerstein's[5] value of $\Delta S=2.7$ J (g-at. °K)$^{-1}$, we get $dT_t/dP=2.2\times10^{-4}$ °K cm^2/kg; this is in poor agreement with the value $dT/dP=9\times10^{-4}$ directly obtained by Grazhdankina.[26] However, a careful examination of the original data presented in that work shows that a value of dT/dP as low as $\sim3\times10^{-4}$ can not be rigorously excluded. The remaining discrepancy could easily result from the possibility mentioned before that the $\Delta L/L$ measurement on a bulk polycrystalline sample may not give an accurate result for $\Delta V/V$.

One further aspect of this transition warrants mention: If it indeed results from the interaction of the exchange and lattice constant, the decreasing deformability of the lattice with increasing pressure should eventually eliminate the first-order character at high enough pressure. This would imply a phase-separation line which simply "stops" in the T-P plane; a situation which is unusual in solids. This elimination of the first-order transition with pressure can also explain the strong nonlinearity observed in dT_N/dP in the following way: The recent articles[26] on the variation of the Eu magnetic ordering temperature with pressure show that the magnetic transition temperature rises for applied pressures of a few kbar, levels off at about 10–20 kbar, and then decreases slowly at higher pressures. This low-pressure behavior is different from that observed in the heavy rare-earth metals; in these, T_N decreases roughly linearly with pressure over a wide range. It seems reasonable to suggest that the rapid increase in ordering temperature observed at low pressures results from a change in T_t with pressure, and at about 10–20 kbar, the first-order transition gets "squeezed out" by the increasing rigidity of the lattice. Then, above 20 kbar, the "normal" decrease of T_N with pressure is observed.

[28] M. Blume, *Hyperfine Structure and Nuclear Radiations*, edited by E. Matthias and D. A. Shirley (North-Holland Publishing Co., Amsterdam, 1968), p. 911.

B. Critical Parameters

It has been shown, both experimentally and theoretically,[1] that in magnetic systems having a paramagnetic to magnetically ordered state transition which is of second order, the sublattice magnetization M in the vicinity of the critical temperature T_c can be described by the relation

$$M=M_0D(1-T/T_c)^\beta, \tag{1}$$

where D is a reduction factor matching this high-temperature formula onto the 0°K magnetization M_0, and β is called the "critical exponent." There are so far no exact criteria as to the ranges of m and T for which this description is accurate. If we assume that the hf field is directly proportional to the sublattice magnetization, we can write Eq. (1) as

$$H=H_0D(1-T/T_c)^\beta. \tag{2}$$

It should be recognized that the analysis here, in terms of critical-point theory, is of very limited validity because the transition is, in fact, first order. Additionally, in the present case, an analysis of the data in terms of Eq. (2) clearly can only be done for $H\geq0.4H_0$. We have arbitrarily selected values from the data taken between $H=0.4H_0$ and $H=0.5H_0$ and least-squares fitted Eq. (2) to them, and shown the results in Fig. 8 and Table II. For the purposes of comparison, the table shows results obtained when the analysis is done over different temperature ranges. It can be seen that β decreases noticeably as points closer to the transition are chosen. At the T_t itself, the functional

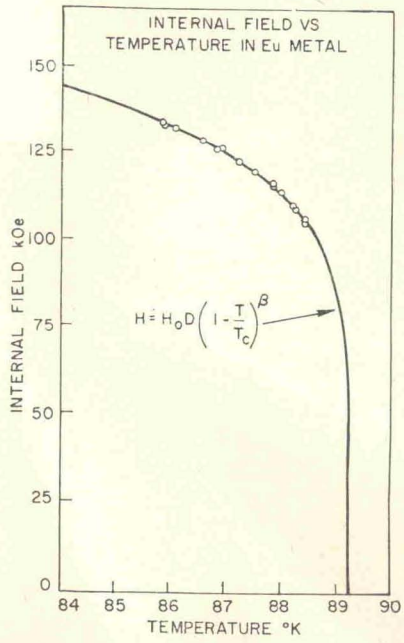

FIG. 8. Fit of the $H=H_0D(1-T/T_c)^\beta$ to hf fields just below $T_t(0.4\leq H/H_0\leq0.5)$. Parameter values obtained from this fit are shown in Table II.

TABLE II. Parameters resulting from least-squares fitting Eq. (2) to the data points over the temperature range (from T_{min} to T_{max}) shown. Only statistical (1σ) errors are shown. Note that the apparent value of β decreases as points closer to the transition are used.

T_{min} (°K)	78.5	78.5	81.5	85
T_{max} (°K)	88.0	88.5	88.5	88.5
β	0.211±0.008	0.198±0.007	0.191±0.009	0.163±0.031
H_0D (kOe)	256±4.2	250±3.3	245±5	224±10
T_N (°K)	89.86 ±0.16	89.58 ±0.10	89.49 ±0.12	89.23 ±0.15

dependence, of course, corresponds to Eq. (2) with $\beta=0$. Although the errors shown in the table appear larger than the variation in β, it must be remembered that the calculations for different temperature ranges were done using many of the same data points, so that the variation of β with temperature range is more significant than the stated error would indicate.

The value for the critical exponent β reported here is rather low and similar values have only been found so far in some low-temperature antiferromagnets ($CuCl_2 \cdot 2H_2O$, $T_c=4.3$°K, $\beta=0.25$ [29]; $CoCl_2 \cdot 6H_2O$, $T_c=2.3$°K, $\beta=0.20$ [30]; DAG, $T_N=2.5$°K, $\beta=0.27\pm0.01$).[31] In most other cases, β seems to have a value in the range 0.3–0.35. The small value of β in our case, presumably, results from the above-reported first-order magnetic transition. Obviously, the interactions which eventually make the transition first order are already effective below the critical temperature and tend to lower the value of β.

In the case of Eu, the existence of the first-order transition makes it obvious that the assumptions implicit in an analysis in terms of critical-point theory are not fulfilled. The interactions producing the first-order behavior in Eu are, in general, present in magnetic systems, though they are not usually strong enough to destroy the continuous nature of the ordering transition. The state of our knowledge is such that we can not, in general, adjust for such perturbations. Thus, any system which appears to have an anomalously low value of β should be tested carefully for the possibility that weak temperature-dependent terms in the magnetic coupling Hamiltonian, rather than a breakdown of critical-point theory, might be responsible.

[29] N. J. Poulis and G. E. G. Hardemann, Physica **19**, 391 (1953), as restated in Ref. 1.

[30] E. Sawatzky and M. Bloom, Can. J. Phys. **42**, 657 (1964); W. van der Lugt and N. J. Poulis, Physica **26**, 917 (1960), as restated in Ref. 1.

[31] J. C. Norwell, W. P. Wolf, L. M. Corliss, J. M. Hastings, and R. Nathans, J. Appl. Phys. **39**, 1232 (1968).

VI. SUMMARY

A careful study of the hf interactions in Eu metal in the temperature regime from 78 to 90°K has been performed by using the Mössbauer effect. At (88.6 ±0.3)°K, the hyperfine field goes from 40% of saturation to zero, indicating a first-order antiferromagnetic to paramagnetic transition in Eu metal at that temperature. Possible reasons for the occurrence of this first-order transition have been discussed and it seems most likely that it is caused by magnetostrictive effects. These effects must involve a lattice distortion similar to that observed in MnO because the isotropic volume change is too small to account for the transition.

In addition, an attempt was made to analyze the temperature dependence of the hf field between $0.4H_0$ and $0.5H_0$ in terms of the equation $H_0D(1-T/T_N)^\beta$. This yielded a value of $\beta=0.16\pm0.04$, which is very much smaller than the usual reported $\beta=\frac{1}{3}$. Because of the relatively high value of H/H_0 and also the proximity of the first-order transition, no further discussion of this low value of β can be made.

ACKNOWLEDGMENTS

We would like to acknowledge technical advice and comments from Dr. G. K. Wertheim concerning the particular problems of measurement, analysis, and discussion of critical-point phenomena. We are greatly indebted to Dr. E. Helfand for theoretical discussions on the comparison of the results with critical-point theory, and to Dr. M. E. Lines for helpful discussions on magnetostrictive first-order transitions. We performed the thermal-expansion measurements using equipment belonging to Dr. E. Fawcett, and we would like to thank him both for making the apparatus available and for helpful comments on the analysis of the data. The approach in Sec. V reflects comments and suggestions by Dr. M. Blume.