Critical Magnetic Properties and Exchange Interactions in EuO†

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The critical magnetic exponents are determined for EuO and found to be $\beta = 0.368 \pm 0.005$, $\delta = 4.46 \pm 0.1$, and $\gamma = 1.29 \pm 0.01$. Within experimental error these values are consistent with the predictions of scaling theory. It is shown that the reduced magnetic field is expressible as a function only of the reduced magnetization in the critical region, with a functional dependence which differs for T above and below T_c. However, the linear model of Schofield *et al.* leads to a single equation of state which gives a good approximation to our data throughout the entire critical region. Our results were analyzed in terms of high-temperature expansion for fcc lattices with nearest J_1 and next-nearest-neighbor J_2 interactions. It is shown that straightforward extrapolation procedures can lead to serious discrepancies. However, the analysis can be used in conjunction with our susceptibility measurements both immediately above T_c and up to 370 °K and with the observed γ^* behavior to determine the value of the magnetic interactions. Our results indicate that J_2 is ferromagnetic and equal to $(0.5 \pm 0.2)J_1$. The disagreement between this and the previously reported value is discussed.

I. INTRODUCTION

A study has been made of the magnetic properties of EuO in the vicinity of the critical temperature T_c. The sample specimen was prepared from a single crystal of high-resistivity EuO grown from the melt as previously described.[1] The principal impurities were C = 100, K = 50, Ca = 35, Sr = 12, and Te = 12 at. ppm, all others less than 10 at. ppm. The sample was ground into a sphere by the A. D. Jones Optical Company, and the measurements were made with a vibrating-coil magnetometer.[2] The results of this study permit a determination of the critical exponents β, δ, and γ, defined by the equations.

$$\sigma = A\epsilon^{\beta} \quad (T < T_c)\,, \tag{1a}$$

$$H = B\sigma^{\delta} \quad (T = T_c)\,, \tag{1b}$$

$$\chi_0^{-1} = C\epsilon^{\gamma} \quad (T > T_c)\,, \tag{1c}$$

where A, B, and C are proportionality constants, $\epsilon = |T - T_c|/T_c$, σ is the magnetic moment in emu/g, H is the internal magnetic field (applied field minus the demagnetization effect), and χ_0 is the initial susceptibility. The evaluation of the critical exponents β, γ, and δ is presented in Sec. II of this paper, and a comparison between our results and predictions of scaling-law theories is given in Sec. III.

Since EuO is a ferromagnetic semiconductor with the Eu^{2+} ions located on fcc lattice sites, it is an ideal material for study in conjunction with the high-temperature expansion of the Heisenberg Hamiltonian. Such a study, using the known expansion coefficients for an fcc lattice with nearest- and next-nearest-neighbor interactions is carried out in Sec. IV. The results of this study enable us to evaluate the relative strength of the two interactions. They also are used to show that extrapolation procedures have limited reliability in the evaluation of γ as a function of the relative interaction strengths.

Our findings are summarized and discussed in Sec. V. The critical parameters are compared with values determined for other materials, and possible reasons for the large discrepancy between the interaction ratio we obtain and its previously reported value are explored.

II. CRITICAL EXPONENTS

A. $T \leq T_c$

The critical index β was determined by the method proposed by Rayl and Wojtowicz,[3] which utilizes the fact that there is a sharp kink in the magnetization-vs-temperature curve at the temperature T_k, where the applied field H_a is equal to the demagnetizing field NM of the sample in the equation $H = H_a - NM$. The magnetization is constant at temperatures below T_k, but falls off sharply at and above T_k. This method has the advantage that the magnetization is accurately determined at $T = T_k$ by the relationship $M = H_a/N$, where N is the demagnetization factor ($N = \frac{4}{3}\pi$ for a sphere). In addition, this method permits an accurate determination of T_c, since $T_k \to T_c$ in the limit $H_a \to 0$.

The observed kink was extremely sharp in our crystal, occurring within an interval of the order of 0.01 K°, which indicates high sample homogeneity. The locus of T_k as a function of applied field is shown in Fig. 1. Extrapolation of this curve to $H_a = 0$ gives $T_c = (69.59 \pm 0.01)$ K°, which precision applies only to temperature differences; the absolute temperature is uncertain by ±0.25 K°, based

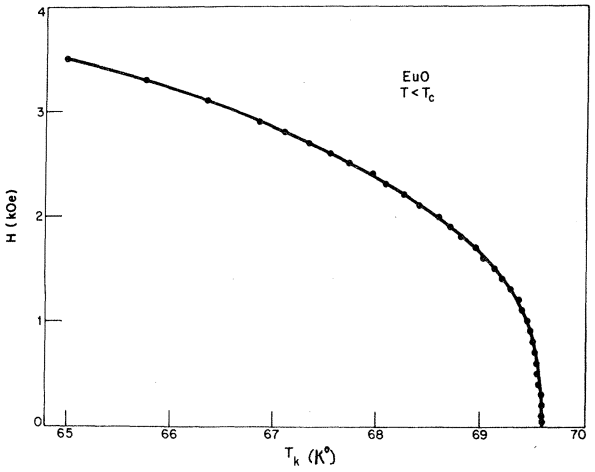

FIG. 1. Experimental relationships between applied field H_a and kink temperature T_k in EuO.

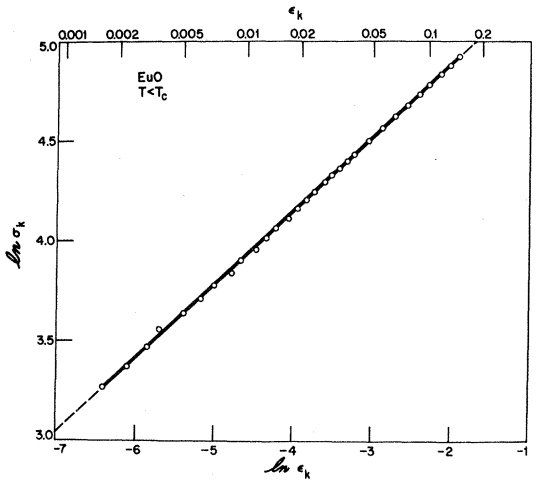

FIG. 2. Kink-point data replotted as σ_k vs ϵ_k on a log-log scale. The solid line corresponds to $\beta = 0.368$, $A = 276$.

on a calibration of our thermocouple at liquid nitrogen. The results shown in Fig. 1 can also be expressed in terms of σ_k vs ϵ_k, where $\epsilon_k = (T_c - T_k)/T_c$ and $\sigma_k = M/\rho = H_a/N\rho$ with $\rho = 8.190$ g/cm^3, the theoretical density for EuO. A log-log plot of the resultant curve is linear over the range $0.0016 < \epsilon_k < 0.15$, as can be seen from Fig. 2, and yields the values $\beta = 0.368 \pm 0.005$ and $A = 276$ for use in Eq. (1a).

The magnetic moment of EuO as a function of magnetic field was measured at $T = T_c$. The resultant relationship was found to satisfy Eq. (1b), with $\delta = 4.46 \pm 0.10$ and $B = 7.6 \times 10^{-6}$.

B. $T > T_c$

The inverse initial susceptibility χ_0^{-1} was determined in the region immediately above T_c by measuring the magnetic moment as a function of temperature at several applied fields between 1 kOe and 50 Oe, by plotting the results in the form of σ^2-vs-H/σ isotherms, and by extrapolating these isotherms to $\sigma^2 = 0$. The ensuing values of χ_0^{-1} were analyzed by two distinct methods in our determination of the critical exponent γ. The first was the T^* technique introduced by Kouvel and Fisher.[4] This involves the determination of

$$T^* = \chi_0^{-1} \Big/ \frac{d\chi_0^{-1}}{dT}$$

as a function of temperature. If a temperature-dependent effective exponent is defined as $\gamma^*(T) = (T - T_c)/T^*$, then $\gamma^*(T) \to \gamma$ in the limit as $T \to T_c$. A computer program was written to evaluate $\gamma^*(T)$ according to this procedure, and it was found that, for $T_c = 69.59$ K°, the value of γ^* is essentially constant, falling within the range 1.278 ± 0.002 for $T - T_c$ between 0.5 and 4 K°, as shown in Fig. 3.

The assumption of a slightly higher value for T_c leads to a general reduction in γ^*, but with a definite maximum at $T - T_c \simeq 3$ K°. Therefore, $\gamma = 1.28$ can be considered a minimum value. When values of 69.585 and 69.58 K° are assumed for T_c, a definite rise in γ^* is observable at the lowest values of $T - T_c$, as is also shown in Fig. 3. Since these values fall within the uncertainty of our experimental determination of T_c, a most probable value of $\gamma = 1.29 \pm 0.01$ has been chosen. However, it should be noted that a value as high as 1.31 cannot be excluded.

The second method of analysis involved the assumption that γ remains constant over the interval 0.5 K° $< T - T_c < 5$ K°, and the use of a computer to obtain a direct determination of those values for γ and T_c which, on the basis of Eq. (1c), best fit

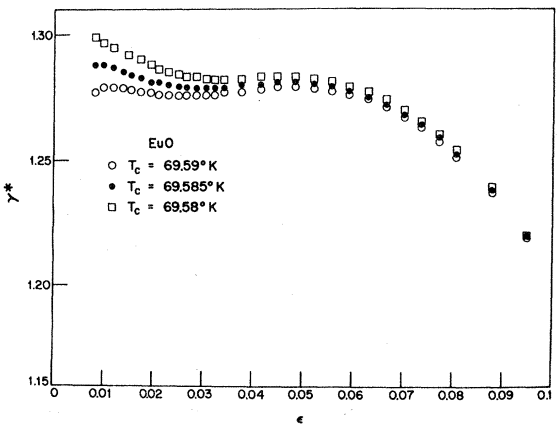

FIG. 3. Experimental values of γ^* as a function of ϵ for a range of assumed values for T_c.

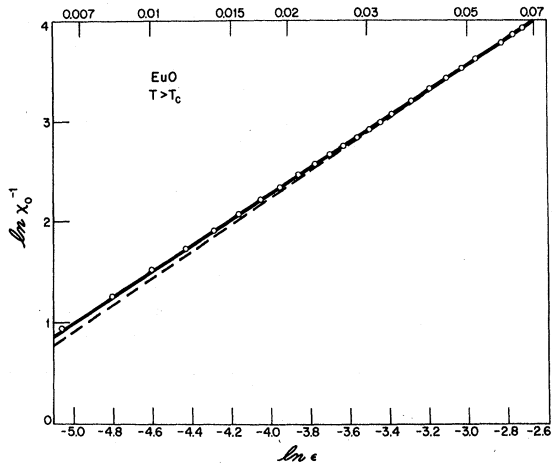

FIG. 4. Experimental values for the inverse initial susceptibility χ_0^{-1} vs ϵ on a log-log scale. The solid line corresponds to $\gamma = 1.285$, $C = 1680$. The theoretical curve for $\alpha = 0.7$ assuming a linear extension is shown by the dashed line and illustrates the poor agreement with experiment for small ϵ, as discussed in Sec. III.

our measurement of χ_0^{-1}. This method yields $\gamma = 1.285$ and $C = 1.68 \times 10^3$ with $T_c = 69.594$ K°. All the values of χ_0^{-1} computed from Eq. (1c) using these values of γ and T_c fell within the experimental error of the measured values. The nature of the agreement is indicated by the logarithmic plot of χ_0^{-1} vs ϵ given in Fig. 4. This value for γ is consistent with the value chosen on the basis of the T^* analysis.

III. SCALING AND EQUATIONS OF STATE

Studies of the magnetic critical-point exponents have led various authors[5-8] to propose the relationship

$$\gamma = \beta(\delta - 1). \tag{2}$$

Equation (2) cannot be rigorously derived, but is based on plausibility arguments. The ultimate check on its validity must come from experimental verification, and is seen to hold in our results, which predict $\gamma = 1.27 \pm 0.05$ on the basis of our measurements of β and δ.

Taking the behavior of the specific heat C_v in the vicinity of T_c as $C_v \propto \epsilon^{-\alpha'}$ for $T > T_c$ and $C_v \propto \epsilon^{-\alpha}$ for $T < T_c$, the scaling laws suggest the additional relationships:

$$\alpha = 2 - \beta(\delta + 1) \tag{3}$$

and

$$\alpha = \alpha'. \tag{4}$$

Our results and Eq. (3) yield $\alpha = 0$ for EuO within our experimental error, in which case the predicted behavior for the specific heat is $C_v \propto \ln\epsilon$.[8] The specific heat of EuO has been investigated by Teaney[9] and a logarithmic relationship observed for $T > T_c$. However, this agreement must be qualified by the fact that different specific-heat behavior is observed for $T < T_c$, indicating that Eq. (4) is not generally valid.

The scaling laws also lead to the prediction[5,8] that the reduced field h is a function of the reduced magnetization m only; i.e., $h = h(m)$, where $h = H\epsilon^{-\beta\delta}$ and $m = \sigma\epsilon^{-\beta}$. Following the nomenclature of Ho and Litster,[10] we have taken $h = \hat{H}\epsilon^{-\beta\delta}$, where $\hat{H} = (g\mu_B/SkT)H$, and have plotted h vs m at various temperatures, as shown in Fig. 5. The resultant points are seen to lie on two curves, one for $T < T_c$, the other for $T > T_c$. These curves are qualitatively similar to those obtained by Ho and Litster[10] for the ferromagnetic insulator $CrBr_3$, which they showed to be consistent with the scaling laws.[8]

It is of considerable interest to establish if a convenient parametric form for the equation of state in the vicinity of T_c can be obtained which is applicable both above and below T_c. One such form, called the linear model (LM), was recently proposed by Schofield, Litster, and Ho.[11] The LM involves transformation to new variables θ and r defined by the equations[12]

$$\hat{H} = ar^{\beta\delta}\theta(1 - \theta^2), \quad \epsilon = r(1 - b^2\theta^2), \quad \sigma = r^{\beta}m(\theta), \tag{5}$$

with

$$b^2 = (\delta - 3)/(\delta - 1)(1 - 2\beta)$$

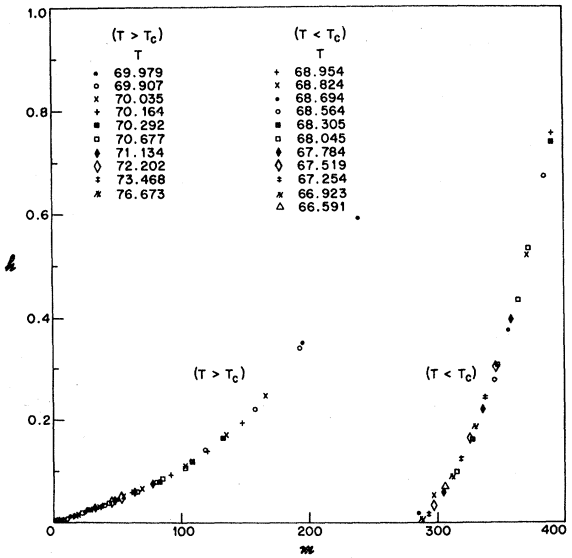

FIG. 5. Reduced magnetic field h plotted as a function of the reduced magnetization m in the critical region both above and below T_c.

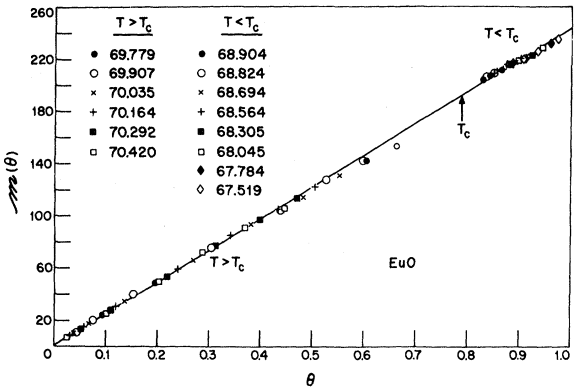

FIG. 6. Comparison of experiment with the linear model for $a = 0.22$ and $b^2 = 1.598$. The straight line corresponds to $m(\theta) = 244\theta$.

and a an arbitrary parameter. The LM then predicts the linear relationship $m(\theta) = k\theta$, where k is a constant. A plot of $m(\theta)$ vs θ for our EuO data taken both above and below T_c is shown in Fig. 6, wherein $a = 0.22$ and $b^2 = 1.598$. A single straight line with $k = 244$ is seen to represent a good approximation to the data both above and below T_c. However, there is a distinct departure from linearity in the region $\theta > 0.5$ and $T > T_c$, with the worst point departing about 5% from the straight-line relationship. In view of the large number of experimental parameters (i.e., σ, H_a, N, ϵ, δ, γ) which enter into the determination of each point, the departure from linearity cannot be considered outside possible experimental error.

A necessary condition for the validity of the LM[11] is

$$BA^{(\delta-1)}/C = b^{(\delta-3)}/(b^2-1)^{(\gamma-1)}, \tag{6}$$

where A, B and C are the coefficients defined in Eqs. (1). Using the experimental values given in Sec. II, and considering only the uncertainty in the value of δ in setting the limits, evaluation of the left-hand side of Eq. (6) gives

$$BA^{\gamma-1}/C = 1.26^{+0.95}_{-0.54},$$

and the right-hand side yields

$$b^{\delta-3}/(b^2-1)^{\gamma-1} = 1.64 \pm 0.05.$$

Thus, using the experimental value of δ, Eq. (6) does not hold, and departure from the LM is to be expected. However, the extreme sensitivity of the left-hand side to small changes in δ places Eq. (6) well within experimental uncertainty. In fact, the evaluation of δ from Eq. (2) using the experimental values for β and γ yields 1.63 for the left-hand side of Eq. (6).

Given these considerations and the fit shown in Fig. 6, it is felt that the LM adequately describes the properties of EuO, and gives a good approximation to the equation of state throughout the critical-temperature region.

IV. USE OF HIGH-TEMPERATURE EXPANSION TO DETERMINE EXCHANGE CONSTANTS

EuO is both semiconducting and ferromagnetic, and hence can be considered a "Heisenberg" ferromagnet. In addition, it has a simple rocksalt structure with the Eu^{2+} ions on an fcc lattice. In the case of a Heisenberg Hamiltonian with nearest- and next-nearest-neighbor exchange interactions J_1 and J_2, respectively, the magnetic susceptibility can be expressed in the form of the power series

$$\frac{T\chi_0}{C} = 1 + \sum_{n=1}^{\infty} a_n(\alpha, S)\left(\frac{J_1}{kT}\right)^n, \tag{7}$$

where $\alpha = J_2/J_1$, and C is the Curie constant. For a material with magnetic ions on an fcc lattice, the coefficients $a_n(\alpha, S)$ have been evaluated for arbitrary spin S through the sixth order.[13–16] EuO is therefore an ideal material in which to investigate exchange interactions by comparing experimental susceptibility measurements with the predictions of Eq. (7).

In addition, the assumption of a power-law divergence of the susceptibility upon approaching T_c from above leads to definite relationships between the critical exponent γ and the coefficients $a_n(\alpha, S)$. Since the power-law assumption is restricted to the limit $T \to T_c$, evaluation of γ as a function of α involves the extrapolation of $a_n(\alpha, S)$ to $n \to \infty (1/n \to 0)$, as discussed in Appendix A. A significant body of literature has been devoted to extrapolation procedures for dealing with this problem. In the past, such calculations have been unencumbered by being subjected to direct experimental verification. Hence the reliability of the extrapolation has generally been assumed on the basis of the behavior of the extrapolated function. However, in EuO we are afforded a rare opportunity to experimentally check the reliability of such an extrapolation.

A. $T \gg T_c$

It is desirable to make comparisons between Eq. (7) and experimental inverse susceptibility in the high-temperature range since the theoretical susceptibility is then much less sensitive to the extrapolated terms than it is immediately above T_c. We therefore measured the susceptibility of our EuO sample at temperatures up to 370 K° in a field of 10 kOe. The resultant χ_M^{-1}-vs-T curve appears linear above 210 K° with a molar Curie constant $C = 7.4$ and an apparent paramagnetic Curie point $\theta = 81$ K°, in fair agreement with the results obtained by McGuire *et al.*[17]

If the measured θ is equated with the molecular-

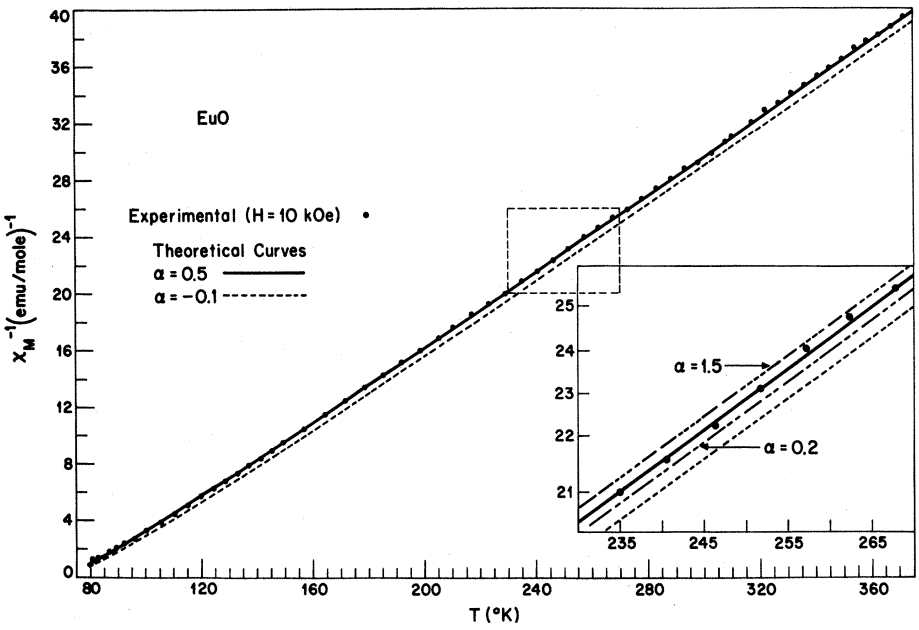

FIG. 7. Inverse susceptibility vs temperature. The experimental values (indicated by the solid dots) were obtained from measurements in a field of 10 kOe by assuming field independence of the susceptibility, which is invalid at the lower temperatures. The solid curve was computed from the high-temperature expansion for an fcc lattice with $S=\frac{7}{2}$ and an exchange ratio $\alpha=0.5$. Similar curves computed for other values of α are also shown for the purpose of comparison.

field Curie point T_M, a value $t_c = T_c/T_M = 0.86$ is obtained. This is greater than the maximum attainable value of t_c. The error lies in equating θ and T_M, i.e., in assuming that, over the temperature interval $3T_c < T < 370$ K°, one can use the equation $\chi = C/(T-\theta)$ as a reasonable approximation to Eq. (7). It can be shown that much higher temperatures are required before the experimental value of θ closely approximates T_M.[18]

This effect is illustrated in Fig. 7 which contains our experimental values of χ_M^{-1} vs T as well as theoretical curves which were calculated for $T_c = 69.59$ K° using a high-temperature series obtained by linear extension of the function described in Appendix A and were scaled to match the experimental Curie constant. The theoretical curve for $\alpha = 0.5$ yields the best fit to the data. Despite appearances, this curve is not a straight line and, if extended to sufficiently high temperatures, is found to correspond to $T_M = 84.05$ K°. For higher values of α the theoretical curves are relatively insensitive to changes in α, but the one for $\alpha = 1.5$ lies about 2 K° below the experimental results, which is outside experimental error. The theoretical curves are more sensitive to changes in α at smaller α values, and by $\alpha = 0.2$ lie more than 2 K° above the experiment; for $\alpha = -0.1$ there is a 5 K° difference. This is in direct contradiction with the results obtained from an NMR experiment by Boyd[19] and from specific-heat measurements by Henderson *et al.*,[20] who reported values of $\alpha = -0.13$ and $\alpha = -0.11$, respectively.

B. T near T_c: Standard Extrapolation

In the temperature region immediately above T_c, the higher-order terms of the high-temperature expansion ($n \geq 7$) make a significant contribution to the susceptibility. Therefore, interpretation of the susceptibility data in this temperature region requires extrapolation from the known coefficients to obtain information about the higher-order terms. Furthermore, as noted above, the experimentally determined value of γ can be related to the magnetic interactions only by extrapolation of Eq. (7) to $n \to \infty$.

In order to perform such an extrapolation, we initially followed the "slope"-method procedure proposed by Stanley.[21] This method, which utilizes the convergence criterion

$$\rho_n \equiv a_n/a_1 a_{n-1} \xrightarrow[n\to\infty]{} T_c/T_M \equiv t_c, \tag{8}$$

defines a function of $1/n$ which extrapolates to $\gamma(\alpha)$ as $1/n \to 0$ and is (hopefully) smoothly varying. Application of this procedure to the fcc lattice with both nearest- and next-nearest-neighbor interactions failed to yield a function smooth enough to extrapolate for $\alpha > 0.4$.

This result is not surprising since, as α becomes large and positive, the next nearest neighbors start to play a dominant role. These neighbors are located on a simple cubic lattice, and it is known[21] that with this structure the extrapolation is less reliable and is best achieved by considering the coefficients in two separate groups, i.e., a_2, a_4, a_6, . . . and a_1, a_3, a_5, ... We therefore substituted a new convergence criterion involving a_{n+1}, a_{n-1} for that given in Eq. (8) and extended the slope method to consideration of this case. This leads to a determination of the function $\gamma_{n+1/2,n-1/2}$ which is defined in Appendix A and is shown plotted vs $1/n$ in

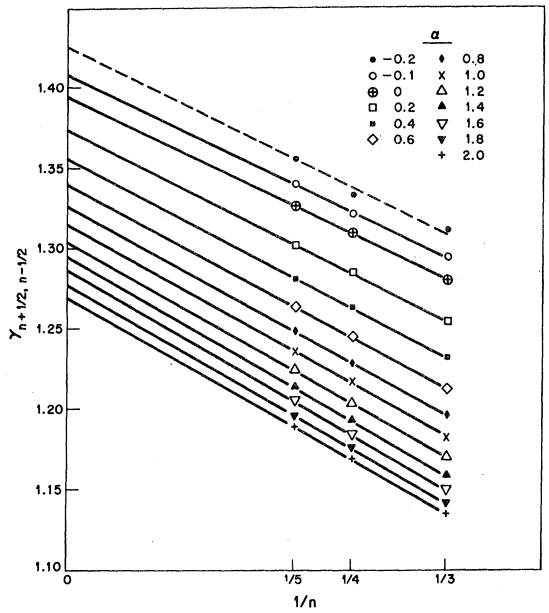

FIG. 8. Function $\gamma_{n+1/2,n-1/2}$ for an fcc lattice with $S=\frac{1}{2}$ plotted against n^{-1} for various values of the exchange ratio α.

Fig. 8. It is seen that $\gamma_{n+1/2,n-1/2}$ varies linearly with $1/n$ over the range $-0.1<\alpha<2.0$. Linear extrapolation to $1/n=0$ therefore leads to a determination of $\gamma(\alpha)$ with an accuracy as good as can be achieved by extrapolation techniques. The series convergence for $\alpha<-0.1$ is poor, which confirms previous findings.[22]

In addition, one can extrapolate the $t_{n+1/2,n-1/2}$ function defined in Appendix A to obtain t_c. However, the t and γ functions are not independent, and it is shown in Appendix B that the linear relationship $\gamma_{n+1/2,n-1/2}=\gamma-a/n$ requires that

$$t_{n+1/2,n-1/2}=t_c(1+a/2n^2+\cdots)\,. \tag{9}$$

Thus linear extensions of the $\gamma(\alpha)$ in Fig. 8 determine, by virtue of Eq. (9), unique extrapolations of the $t_c(\alpha)$. The resulting values of γ and t_c are shown plotted vs α in Fig. 9.

The experimental value $\gamma=1.29\pm0.01$ corresponds in Fig. 9 to the value $\alpha=1.5\pm0.2$, in clear disagreement with the high-temperature susceptibility measurements as shown in Fig. 7. Since the high-temperature susceptibility depends primarily on the known coefficients and hence is relatively insensitive to extrapolation effects, this discrepancy raises serious questions regarding the reliability of extrapolation procedures for the determination of γ.

To investigate this aspect of the problem, we have compared the experimental values of χ_0^{-1} immediately above T_c, as discussed in Sec. II, with the theoretical curves for several values of α. In this temperature interval, as noted previously, the theoretical susceptibility curves will depend strongly on the extrapolated terms. Use of the linear extension (LE) of $\gamma_{n+1/2,n-1/2}$ leads to the curves shown in Fig. 10. It is seen that $\alpha=1.5$, which corresponds to the correct γ value, gives poor agreement with the susceptibility, as does $\gamma=-0.1$. The best agreement with the magnitude of χ_0^{-1} occurs for $\alpha=0.7$. However, the γ value corresponding to this curve is 1.333 which is well outside experimental uncertainty. Hence, the apparently excellent agreement, as indicated by the curve, is misleading. It arises from the fact that the greatest percentage error between the experimental and theoretical curve occurs for T near T_c, where the magnitude of χ_0^{-1} is small. There is almost a 5% error between the experimental results and the theoretical curve for $\alpha=0.7$ at $T-T_c=1\,\mathrm{K}^\circ$, increasing to about 8% by $T-T_c=0.5\,\mathrm{K}^\circ$. This increased deviation as $T\to T_c$ is more clearly illustrated in Fig. 4 where the theoretical curve is given by the dashed line. Thus, one cannot achieve consistency with all our experimental results using a straightforward linear extrapolation of $\gamma_{n+1/2,n-1/2}$ to define a_n for $n>6$.

Attribution of this inconsistency to the unreliability of the linear extrapolation can be checked directly by treating the theoretical inverse-susceptibility curves as data, performing a T^* analysis, and comparing the resultant γ^* values with those obtained from the experiment. As seen in Fig. 11, the theoretical values of γ^* for $\alpha=1.4$ (corresponding to $\gamma=1.294$) fall off with increasing temperature significantly more rapidly than is found experimentally. We must therefore conclude that the extrapolation procedure is incapable of predicting γ to the close tolerances required to make it useful for determining magnetic interaction parameters.

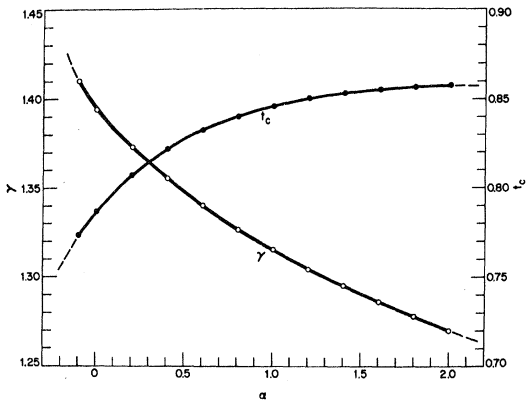

FIG. 9. Extrapolated values for γ and t_c as functions of the exchange ratio γ based on the linear-extension procedure.

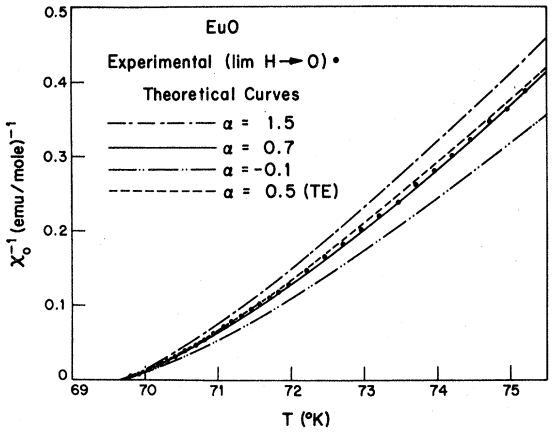

FIG. 10. Comparison between experimental values for the inverse initial susceptibility immediately above T_c and curves computed from the high-temperature expansion for an fcc lattice with $S=\frac{7}{2}$ and various values of the exchange ratios α. The curves for $\alpha=1.5$, 0.7, and -0.1 are based on the linear extension of $\gamma_{n+1/2,n-1/2}$, the curve for $\alpha=0.5$ is based on the truncated extension.

C. T near T_c: Corrected Extrapolation

In order to achieve consistency between theory and the experiments it is necessary that the first six coefficients a_n ($n \leqslant 6$) be those for $\alpha \cong 0.5$ to agree with the high-temperature susceptibility curve, and that the corresponding $\gamma_{n+1/2,n-1/2}$ curve extrapolate to $\gamma \cong 1.29$ as $1/n \to 0$. It is further necessary that the resultant high-temperature expansion series lead to a theoretical χ_0^{-1}-vs-T curve whose magnitude agrees with experiment immediately above T_c, and that the theoretical γ^*-vs-ϵ curve be slowly varying in this temperature interval to ensure agreement in the shape of the susceptibility curve.

The simplest way to meet the first pair of criteria is to linearly extend the $\gamma_{n+1/2,n-1/2}$ corresponding to the known coefficients a_n ($n \leqslant 6$) for $\alpha = 0.5$ until the value $\gamma_{max}=1.29$ is attained, and then to maintain this value as $1/n \to 0$, with the $t_{n+1/2,n-1/2}$ being determined consistently. This type of extrapolation will be referred to as the truncated extension (TE), to distinguish between it and the LE.

Since the high-order terms of Eq. (7) play an important role, lowering the value of $\gamma_{n+1/2,n-1/2}$ in the extended region strongly affects the theoretical χ_0^{-1} curve immediately above T_c. In general, it raises the value of χ_0^{-1} for a given value of α. For the extreme case of the TE, the theoretical curve for $\alpha=0.5$ gives the agreement shown in Fig. 10. In addition, the theoretical curve for $\alpha=0.5$ with the TE leads to the γ^*-vs-ϵ behavior shown in Fig. 11, which is seen to be reasonably consistent with the experimentally observed behavior.

In the high-temperature region ($2T_c < T < 370$ K°), the higher-order terms ($n>6$) play a minor role in the determination of χ. Thus the theoretical χ^{-1}-vs-T curve in this temperature range for a particular value of α is virtually independent of the nature of the extrapolation chosen, and the conclusions reached in connection with Fig. 7 remain unchanged. Thus the high-temperature expansion for $\alpha=0.5$ yields consistent agreement with the experimental value of γ, with the γ^*-vs-ϵ behavior, and with the inverse-susceptibility curves both immediately above T_c and in the region of higher temperatures, provided the TE is used.

The consistency of agreement between theory and experiment is lost when α departs significantly from the value 0.5. Although the magnitude of the theoretical inverse susceptibility is in good agreement with experiment for $\alpha=0.7$, except in the immediate vicinity of T_c when using the LE, reducing the extrapolation to meet the experimental value of γ must have the effect of raising the theoretical curve. For $\alpha=0.7$ and using the TE, the theoretical curve lies ~3% above the experimental results. Therefore $\alpha=0.7$ can be considered the upper bound for α. At the other end, it is seen in Fig. 8 that for $\alpha=0.3$, the value of $\gamma_{n+1/2,n-1/2}$ has reached 1.29 for $n=5$. A lower value of α would require that the extrapolation go through a maximum and then decrease to $\gamma=1.29$ at $1/n=0$. Thus, one would reasonably expect $\alpha>0.3$. Direct confirmation comes from the high-temperature inverse-susceptibility results, where it was shown that by $\alpha=0.2$ the theoretical curve lies 2 K° above the experimental values, with the discrepancy increasing with decreasing α. For these reasons,

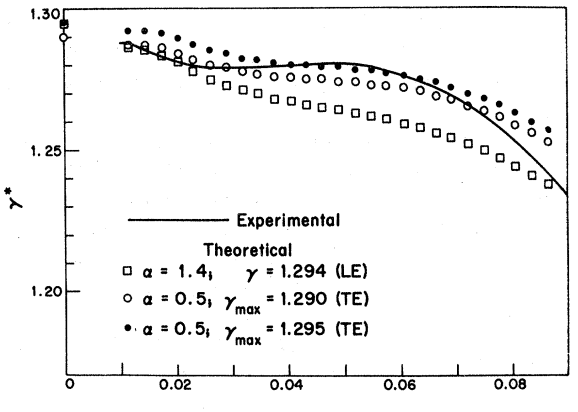

FIG. 11. Experimental and theoretical variation of γ^* with ϵ. For the LE, reduction of α causes a pronounced vertical displacement of the curve. For TE, the position of the curve depends more on γ_{max} than on α. The experimental curve used for comparison corresponds to the solid dots of Fig. 3.

we conclude that the value of α can be bracketed as $\alpha = 0.5 \pm 0.2$. The corresponding value for the nearest-neighbor constant is $J_1/k = (0.53 \pm 0.05)\,\mathrm{K}^\circ$.

V. SUMMARY AND DISCUSSION

We have investigated the magnetic properties of EuO in the vicinity of the Curie point and determined the critical exponents $\beta = 0.368 \pm 0.005$, $\delta = 4.46 \pm 0.10$, and $\gamma = 1.29 \pm 0.01$. The relationship of these values agrees, within experimental error, with the predictions of scaling theory. They are also consistent with the logarithmic behavior of the specific heat observed[9] in EuO immediately above T_c. In further agreement with scaling theory, it was found that for $h = H\epsilon^{-\beta\delta}$ and $m = \sigma\epsilon^{-\beta}$, h can be expressed as a function of m alone. However, the form of $h(m)$ was different for T above and below T_c. An attempt was made to see if a single equation of state could be established for EuO which was applicable both above and below T_c. It was found that the linear model recently proposed by Schofield *et al.*[11] represented at least a good approximation throughout the critical-temperature region.

In addition, our results were analyzed in terms of the high-temperature series expansion for fcc lattices with nearest- and next-nearest-neighbor interactions. On the basis of the known expansion coefficients and susceptibility measurements in the high-temperature region where the results are relatively insensitive to extrapolative procedures, we determined $\alpha \cong 0.5$. We were then able to use this result to test the reliability of extrapolation for the determination of γ.

Griffiths[23] has recently published an article which postulates that in a Heisenberg ferromagnet the critical exponents remain constant, independent of distant-neighbor interactions. This is not borne out by our numerical evaluations of the first six coefficients which show $\gamma_{n+1/2,n-1/2}$ to be strongly dependent on the interaction ratio α for these known terms.

Using a modification of the slope method, we succeeded in obtaining a well-behaved linear relationship between $\gamma_{n+1/2,n-1/2}$ and $1/n$ for $n = 3$, 4, and 5, and found that a straightforward linear extension to $1/n = 0$ gave $\alpha = 1.5$ for the measured value of γ. The linear extension also predicted a much more rapid variation of γ^* vs ϵ than is observed experimentally. The threefold error in α is found to correspond to an error in the γ extrapolation of approximately $4\frac{1}{2}\%$. We believe this error represents the limit of reliability inherent in the determination of γ by extrapolation, since the linear dependence of the known $\gamma_{n+1/2,n-1/2}$ upon $1/n$ leaves negligible uncertainty in their extension. This large unreliability is not surprising when one considers that the extrapolation of γ from the changing slopes $\gamma_{n+1/2,n-1/2}$ involves the second derivative of the ratio $\sigma_{n+1/2}$ with respect to $(n+\frac{1}{2})^{-1}$; the extrapolated value of t_c depends predominantly on the first derivative of these ratios and remains reliable to better than 1%. Given the rapid variation of α with γ (Fig. 9), an unreliability factor of $4\frac{1}{2}\%$ renders this technique meaningless for α determinations.

By using the TE to join the known expansion coefficients for $\alpha = 0.5$ with the experimental value of γ, we were able to obtain reasonable agreement with the shape of the susceptibility curve immediately above T_c (as determined by the γ^*-vs-ϵ curve) and with the susceptibility magnitude both immediately above T_c and in the high-temperature region. This consistency between theory and experiment was limited to the range of values $\alpha = 0.5 \pm 0.2$,[24] corresponding to $J_1/k = (0.53 \pm 0.005)\ \mathrm{K}^\circ$.

The previously reported[19,20] value of $\alpha \cong -0.12$ is clearly incompatible with any of our results. Although the measurements of Boyd[19] and of Henderson *et al.*[20] were conducted at or below 4.2 K° while ours were near or above T_c, it is unreasonable to attribute such a large discrepancy to a temperature variation of the exchange parameters caused by a change in lattice dimensions.[25] Direct evidence to the contrary is provided by measurements of the pressure dependence of T_c, which show dT_c/dP to be small and positive in EuO.[26–28]

The NMR results were obtained[19] by parametrizing the magnetic properties of EuO in terms of the near-neighbor exchange interaction J_1/k, the exchange ratio α, the magnetization M, and an effective field. Curves of $M(0) - M(T)$ vs T for various values of the parameters were then calculated on the basis of spin-wave theory as given by Charap and Boyd,[29] and compared with the measured values of $f(0) - f(T)$ vs T, where $f(T)$ is the NMR frequency at temperature T, and $f(0)$ is obtained by extrapolation to $T = 0$. A "best fit" was then obtained for $\alpha = -0.130 \pm 0.005$. However, in a similar experiment on EuS,[29] it was noted that for all the values considered ($-0.6 < \alpha < 0.3$) it was possible to find a value of $H(>0)$ which produced a set of values of J_1/k such that the rms deviation was of the order of 1%. No such discussion was given for EuO, and it would be of interest to know if the value of the parameters J_1/k and α obtained here are consistent with the NMR results for some positive value of H.

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APPENDIX A

It is convenient to rewrite Eq. (7) as

$$\frac{T\chi_0}{C} = 1 + \sum_{n=1}^{\infty} a_n'(\alpha, S)\left(\frac{T_M}{T}\right)^n , \tag{A1}$$

where $a_n' = a_n/a_1^n$, since $a_1(\alpha, S)J_1 = kT_M$. Considering the convergence criterion in terms of alternate coefficients defines a new function

$$\sigma_{n+1/2} \equiv (a_{n+1}'/a_{n-1}')^{1/2} \xrightarrow[n\to\infty]{} T_c/T_M \equiv t_c . \tag{A2}$$

The assumption of a power-law divergence of susceptibility upon approaching T_c from above requires that

$$\frac{T\chi_0}{C} \propto \left(1 - \frac{T_c}{T}\right)^{-\gamma} \tag{A3}$$

as $T \to T_c$. The right-hand side of Eq. (A3) can be expanded as a power series in T_c/T. Equating the coefficients of corresponding powers of T^{-n} in Eqs. (A1) and (A3) leads to

$$\sigma_{n+1/2} = t_c\left[\left(\frac{\gamma+n}{n+1}\right)\left(\frac{\gamma+n-1}{n}\right)\right]^{1/2} , \tag{A4}$$

which can be written as

$$\sigma_{n+1/2} = t_c\left[\left(1 + \frac{(\gamma-1)(n+\frac{1}{2})}{n(n+1)}\right)^2 - \left(\frac{\gamma-1}{2n(n+1)}\right)^2\right]^{1/2}$$

$$= t_c\left(1 + \frac{(\gamma-1)(n+\frac{1}{2})}{n(n+1)}\right)(1 - \tfrac{1}{2}X^2 - \tfrac{1}{8}X^4 - \cdots) ,$$

where

$$X = (\gamma-1)/[2n(n+1) + (\gamma-1)(2n+1)].$$

Taking $\gamma = 1.3$, we note that for $n=2$, $X^2 \cong 0.0005$ and for $n=3$, $X^2 \cong 0.0001$. Therefore, to an excellent approximation,

$$\sigma_{n+1/2} \cong t_c[1 + (\gamma-1)(n+\tfrac{1}{2})/n(n+1)] \tag{A5}$$

for such a divergence. The assumption of power-law behavior only in the limit $T \to T_c$ still requires that this expression be valid for sufficiently large n. Therefore, the slope of the plot of successive values of $\sigma_{n+1/2}$ vs $1/(n+\frac{1}{2})$ will become equal to $t_c(\gamma-1)$ in the limit as $1/(n+\frac{1}{2})$ approaches zero.

Using Eq. (A5) in conjunction with the slope method leads to the new functions, completely analogous to those found by Stanley[21] on the basis of Eq. (8),

$$t_{n+1/2,n-1/2} = \tfrac{1}{2}(\sigma_{n+1/2} + \sigma_{n-1/2}) + (n - 1/2n)(\sigma_{n+1/2} - \sigma_{n-1/2}), \tag{A6}$$

$$\gamma_{n+1/2,n-1/2} = 1 - [n(n+1)/n+\tfrac{1}{2}] \times (1 - \sigma_{n+1/2}/t_{n+1/2,\,n-1/2}) , \tag{A7}$$

which approach t_c and γ, respectively, as $n \to \infty$.

APPENDIX B

Equations (A6) and (A7) for $t_{n+1/2,n-1/2}$ and $\gamma_{n+1/2,n-1/2}$, respectively, were obtained by writing out Eq. (A5) for both $\sigma_{n+1/2}$ and $\sigma_{n-1/2}$ and then solving this pair of equations for t_c and γ. Therefore, substitution of $t_{n+1/2,n-1/2}$ and $\gamma_{n+1/2,n-1/2}$ must necessarily satisfy Eq. (A5) for $\sigma_{n+1/2}$. Since similar consideration of the pair of equations for $\sigma_{n+3/2,n+1/2}$ requires that the substitution of $t_{n+3/2,n+1/2}$ and $\gamma_{n+3/2,n+1/2}$ for t_c and γ must also satisfy Eq. (A5) for $\sigma_{n+1/2}$, it follows that

$$\sigma_{n+1/2} = t_{n+1/2,n-1/2}[1 + (\gamma_{n+1/2,n-1/2} - 1)(n+\tfrac{1}{2})/n(n+1)]$$

$$= t_{n+3/2,n+1/2}[1 + (\gamma_{n+3/2,n+1/2} - 1)(n+\tfrac{1}{2})/n(n+1)] . \tag{B1}$$

Assuming a linear extension

$$\gamma_{n+1/2,n-1/2} = \gamma - a/n \tag{B2}$$

and arbitrarily expanding

$$t_{n+1/2,n-1/2} = t_c(1 + b_1/n + b_2/n^2 + \cdots), \tag{B3}$$

it is found on substitution and multiplication by $n(n-1)/(n+\frac{1}{2})$ that

$$0 = t_c\left[\left(1 + \frac{b_1}{n} + \frac{b_2}{n^2} + \cdots\right)\left(\frac{n(n+1)}{n+\frac{1}{2}} + \gamma - 1 - \frac{a}{n}\right)\right.$$

$$\left. - \left(1 + \frac{b_1}{n+1} + \frac{b_2}{(n+1)^2} + \cdots\right)\left(\frac{n(n+1)}{n+\frac{1}{2}} + \gamma - 1 - \frac{a}{n+1}\right)\right]$$

$$= t_c\left[\frac{b_1}{n+\frac{1}{2}} + b_2\frac{n(n+1)}{n+\frac{1}{2}}\left(\frac{1}{n^2} - \frac{1}{(n+1)^2}\right)\right.$$

$$\left. + [b_1(\gamma-1) - a]\left(\frac{1}{n} - \frac{1}{n+1}\right) + \cdots\right]$$

$$= t_c\left(\frac{b_1}{n+\frac{1}{2}} + \frac{[2b_2 - a + b_1(\gamma-1)]}{n(n+1)} + \cdots\right) . \tag{B4}$$

Since the coefficients must vanish for all orders of n, it follows that

$$b_1 = 0 \quad \text{and} \quad b_2 = \tfrac{1}{2}a . \tag{B5}$$

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Cooperative Dynamic Jahn-Teller Effect. II. Crystal Distortion in Perovskites

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A molecular-field-type theory is given for the second-order phase transitions occurring in perovskite crystals, in which a Jahn-Teller (JT) ion (Mn^{3+}, Cu^{2+}, Cr^{2+}) occupies an octahedral B site. The dynamic character of the JT effect is taken into account and excited vibronic states are included. The ordered system consists of two sublattices, each having the same tetragonal, but opposite orthorhombic, mean distortion amplitudes. Near the transition temperature T_t, the two amplitudes behave as $T_t - T$ and $(T_t - T)^{1/2}$, respectively. Increasing the anisotropic JT coupling β enhances the mean tetragonal distortion amplitude and diminishes the orthorhombic one. The transition temperature is studied as a function of the molecular field strength and of β, and characteristic regions of solutions are distinguished. The temperature dependence of the specific heat for some typical systems is presented.

I. INTRODUCTION

In a previous paper[1] (henceforth referred to as I), a theory of cubic-to-tetragonal phase transformations in spinels due to a cooperative Jahn-Teller (JT) effect has been given. Compared to spinels, the experimental material[2–14] concerning JT-induced transformations in perovskitelike compounds is less extensive. On the other hand, in the perovskites there exists rather definite experimental evidence from the anisotropic ESR spectra due to Cu^{2+} in low concentrations,[15] that there are individual distorted JT centers even in the cubic phase.

Perovskites are seldom found to have a simple cubic structure; owing to packing-induced distor-