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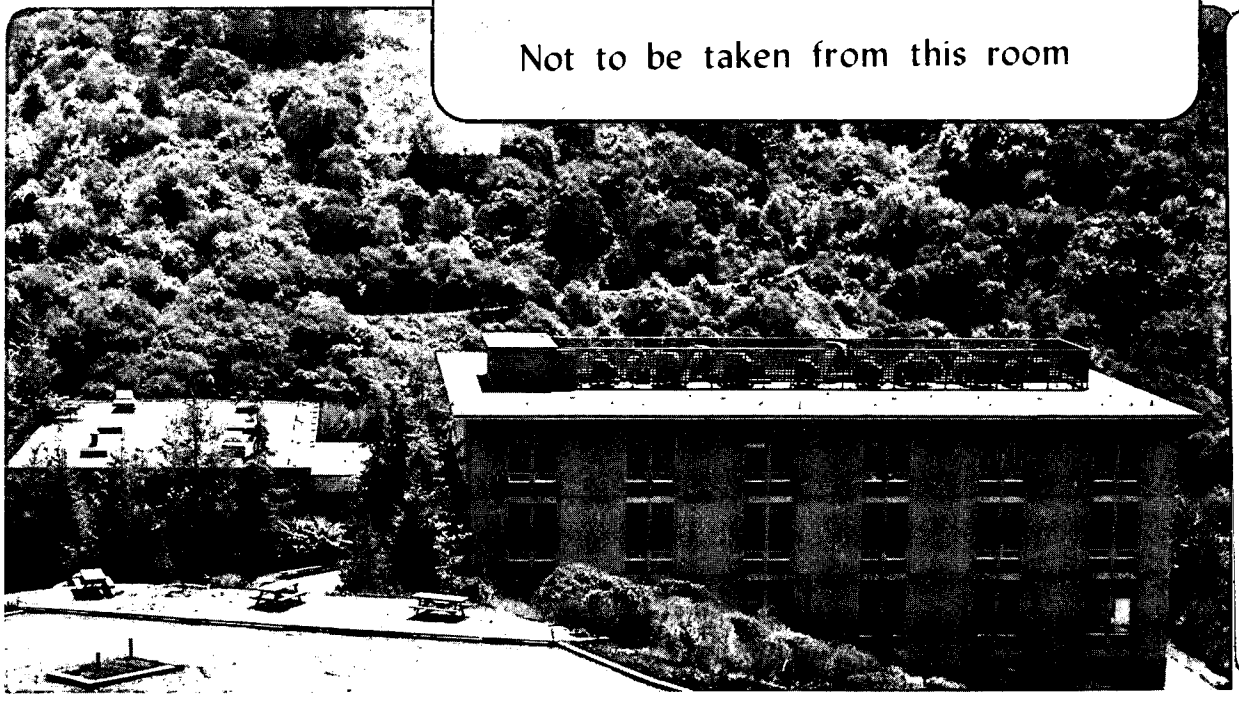

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Specific-heat data in the 0.3 to 20K region for two high-quality UPt_3 samples are analyzed with emphasis on the 4-10K region. It is concluded that no effect associated with antiferromagnetic ordering has been observed, and an upper limit of the order of 0.1% for such an effect is deduced. The results are compared with earlier measurements in the vicinity of the Néel temperature, 5K.

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Antiferromagnetic ordering in UPt_3 is of particular interest in relation to the splitting of the superconducting transition[1] with its implication of unconventional superconductivity and the possibility that the magnetic properties influence the nature of the superconducting transition. Weak antiferromagnetic ordering in UPt_3 at a Néel temperature $T_N \sim 5K$ was first observed in μSR measurements.[2] Subsequent neutron-diffraction measurements have shown the presence of ordered moments with a magnitude of the order of $10^{-2}\mu_B$ /U-atom in some, but not all, UPt_3 samples[3]. As a contribution to establishing a better understanding of the magnetic properties of UPt_3, we report here measurements of the specific heat, C, on two high-quality samples in the vicinity of the Néel temperature. Although the results show qualitative similarities to earlier measurements, including measurements[4] on one of the same samples, they are interpreted differently. In particular, it is concluded that, to within a precision of approximately 0.1%, there is no intrinsic feature in the specific heat near the antiferromagnetic ordering. (In Ref. 4 it was concluded that the anomalies in C near 7K were intrinsic, although probably not associated with the antiferromagnetic ordering.)

Measurements of C on the same two samples in the vicinity of the critical temperature for superconductivity, $T_c \sim 0.5K$, have been reported elsewhere.[1] The samples, number 1 and number 2, are identified by the same numbers here and in Ref. 1, and information on sample preparation and other properties is given in Ref. 1 and in references cited there. Sample 1 was also one of a group of three studied earlier[4] in the vicinity of T_N, but was designated sample 3 in that work.

An overview of the data for samples 1 and 2, in both zero field and 7T is given in

Fig. 1 which also includes zero-field and 8T data for a third sample, sample 3, that was used for studies[5] of the pressure dependence of C. The solid curve in the figure is a fit to the zero-field data for sample 2 to the expression $C = \gamma T + \delta T^3 \, InT + \varepsilon T^3 + B_5 T^5$ but is reproduced with the data for samples 1 and 3 to facilitate comparisons of the data for the different samples. The data for sample 3 were taken on the same temperature scale used for the measurements on samples 1 and 2. They are included for comparison because the temperature dependence and the magnetic field dependence of C are noticeably different from those for samples 1 and 2 illustrating the effect of sample quality on C. They will not be considered in connection with the effects of antiferromagnetic order, however, because the density of data points in the vicinity of T_N was substantially lower and the precision of the data was somewhat lower than for samples 1 and 2.

Between 4 and 10K, all measurements were made with the same high-sensitivity thermometer. The thermometer was shielded from the magnetic field in a way that has been shown to limit the effect of field on the thermometer calibration to less than 0.1% in C. For both samples, in both zero field and 7T, the data show similar structure -- deviations of approximately 0.3% from smooth curves in the 5 to 8K region -- as shown in Fig. 2, where the zero-field data for sample 1 (open circles) are compared with the zero-field data for the same sample from Ref. 4 (solid triangles). In magnitude, the two sets of data are essentially identical, and to facilitate comparison they are plotted separately, shifted relative to each other on the vertical scale, but compared with an identical reference curve in each case. The deviations from the reference curve are qualitatively similar in the two cases, but appreciably smaller in amplitude in this work.

Since the temperature scale used for the measurements reported here is known to include irregularities that can produce spurious structure of the order of several tenths of a percent in C, it is necessary to consider the possibility that the structure apparent in Fig. 2 is related to temperature-scale error and does not reflect an intrinsic property of the sample. One way of distinguishing between these two possibilities is by comparison with the values of C for a reference material such as Cu measured with the same thermometer. Such a test is illustrated in Fig. 3: the expression $C = \gamma T + \delta T^3 \ln T + \varepsilon T^3$, which is known to give a reasonable approximation to C for UPt_3 in this temperature interval, was used to make separate fits to the 4-10K data for each UPt_3 sample in each field; 4-10K, zero-field data for a high-purity Cu sample were also fitted by the 3-parameter expression appropriate to Cu in that temperature interval, $C = \gamma T + B_3T^3 + B_5T^5$. The fractional deviations from each of the 5 fitting expressions are compared as functions of temperature in Fig. 3. The striking similarity of the deviations is convincing evidence that they arise from temperature-scale error rather than intrinsic properties of the samples.

Further evidence bearing on the origin of the structure in C is provided by consideration of the nature of the temperature scale -- an R-T relation that gives the temperature, T, as a function of the resistance, R, of the thermometer -- and its relation to the calculated C. In this case, the temperature scale has two components that correspond to two stages in its development. The first is an R-T equation of the form $T^{-1}=\Sigma A_n(\ln R)^n$, that approximates the calibration data to within a few percent in the 2-30K region. It has 6 terms with coefficients that alternate in sign, and the sign of the deviations from the fit also alternates in sign with changing temperature. The second component is a "difference

curve," actually a tabular representation of a hand-drawn curve through the fractional deviations of the calibration data from the equation, that is used to correct values of T calculated from the equation. Inclusion of the difference curve in the calculation of T provides a significant increase in the overall accuracy of the derived values of C. However, particularly since C depends on the temperature derivative of errors in the temperature scale, i.e., on the slope of the difference curve, errors in the curve that are within the uncertainty of the calibration data can produce small errors in C that change sign in short temperature intervals (comparable to the spacing of the nodes in the difference curve). To demonstrate that the difference curve can produce structure in C similar to that evident in Fig. 3, two calculations, one with and one without a difference curve, of the 4-10K, zero-field data for sample 1 are compared in Fig. 4. As in Fig. 2, the open circles represent the calculation with the complete R-T relation including the difference curve, but in Fig. 4 C/T has been divided by a+bT, which is a linear approximation to C/T in that temperature interval, to permit display on an expanded scale. For comparison the solid squares represent $C^*/T^*(a^*+b^*T^*)$ in which the corresponding quantities are all calculated from a three-term equation that approximates the R-T relation in the 4-10K temperature interval, but without the use of a difference curve. Furthermore, the coefficients of the terms in the R-T equation were forced to be positive to avoid systematic alternation in the sign of the deviations from the fit which could also produce structure in $C^*(T^*)$. On average, the open circles are certainly a better approximation to C, but they do show structure introduced by the difference curve that is not apparent in the solid squares.

On the basis of the measurements reported here, and the comparisons described

above, we conclude that, for both sample 1 and sample 2, there is no structure in C in the 4-10K region that cannot be understood as arising from temperature-scale irregularities. Since sample 1 was also one of a group of three samples that showed essentially identical structure in other measurements[4], and since that structure was qualitatively similar to (although greater in magnitude than) that reported here, we suggest that all of the observed structure in C originates in temperature-scale irregularities. (Given the difficulty in establishing a temperature scale in the 4-10K region, the similarity in the structure in the two independent measurements on sample 1 is not inconsistent with this interpretation. For any scale with practical sensitivity, a transition must be made from vapor pressure thermometry at lower temperatures to gas thermometry at higher temperatures. Futhermore, the virial corrections to gas-thermometer temperatures are substantial in this region, and might introduce similar errors into different scales.) Since both samples 1 and 2 showed antiferromagnetic order[2,6], this interpretation of the data implies an upper limit of approximately 0.1% for the effect of antiferromagnetic ordering on C (see Fig. 3).

It is of some interest to compare the upper limit for the effect of antiferromagnetic ordering on the specific heat of UPt_3 with that observed for the itinerant-electron antiferromagnetic ordering in Cr. In the latter case, there is a clearly defined specific-heat anomaly extending from ~280K to ~315K with a sharp peak near 310K.[7] The associated entropy was estimated[7] to be 20mJ/K mole (but a reanalysis[8] of the same data gave an estimate that was higher by a factor 2). With the assumption that any specific-heat anomaly in UPt_3 is similar in shape to that observed in Cr, the upper limit of 0.1% in C for UPt_3 corresponds to an upper limit of ~0.1 mJ/K mole -- more than two orders of magnitude

smaller than observed for Cr. Although it is not obviously relevant to the entropy comparison, the magnetic moments observed[3] in UPt_3 are smaller, by a factor of approximately 40, than those reported[9] for Cr.

As noted above, and illustrated in Fig. 1, there are significant differences in both the temperature and field dependences of C among the three samples. Samples 1 and 2, for which the temperature dependences of C are essentially the same, are the "better" samples (as reflected, e.g., in the sharpness of the superconducting transition) and most likely to exhibit the properties characteristic of pure UPt_3. For sample 2, the effect of 7T on C is negligible. For both samples 1 and 3 below 7K, C is increased by the application of a field, but the effect is substantially greater for sample 3. The belief that sample 3 is the lowest in quality suggests that the similar field dependences observed for samples 1 and 3, and in particular the relatively large uniform shift of C/T in an 8-T field previously reported[5] for sample 3, are not characteristic of pure UPt_3. It is probable that sample 2 best represents the low-field field dependence of C for UPt_3. In any case, it is unlikely that the field dependences observed are associated with magnetic moments that order near 5K.

The work at Berkeley and Los Alamos was supported by the Director, Office of Energy Research, Office of Basic Energy Science, Division of Materials Sciences of the U.S. Department of Energy (at Berkeley under contract No. DE-AC03-76SF00098).

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Figure Captions

Fig. 1. Specific-heat data for three UPt_3 samples. Zero-field points for $T<T_c$ are omitted. The solid curves are identical, included only to illustrate sample-to-sample differences.

Fig. 2. Two independent zero-field measurements of C for sample 1: ○, this work; Δ, Ref. 4.

Fig. 3. Percentage deviations from appropriate 3-parameter fits (see text) to the data for samples 1 and 2, and for a high purity Cu sample.

Fig. 4. Two different calculations of the same zero-field data for sample 1: ○, with the complete R-T temperature-scale relation; □ with a simplified R-T equation and no difference curve. See text for details.

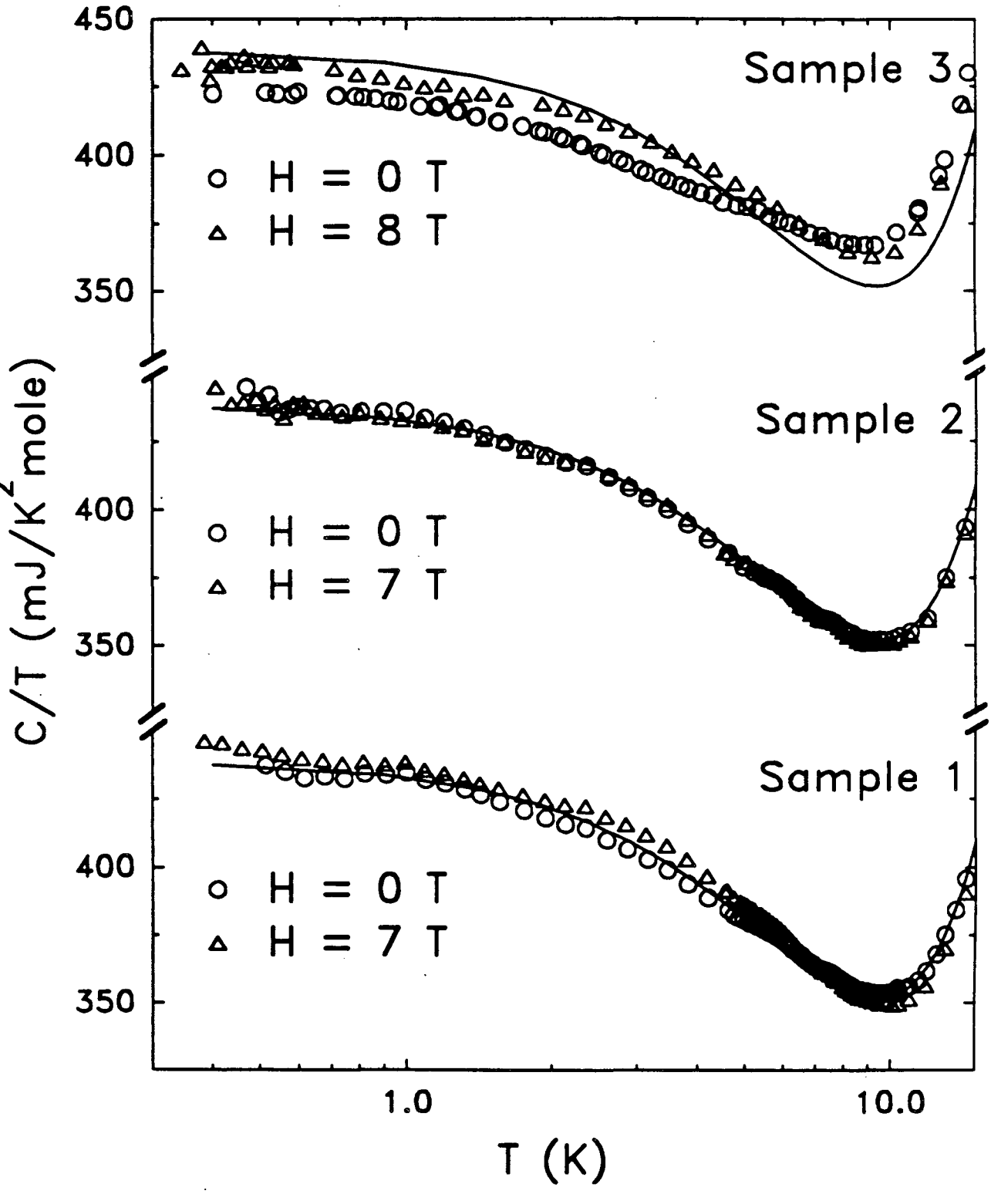

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FIG. 1

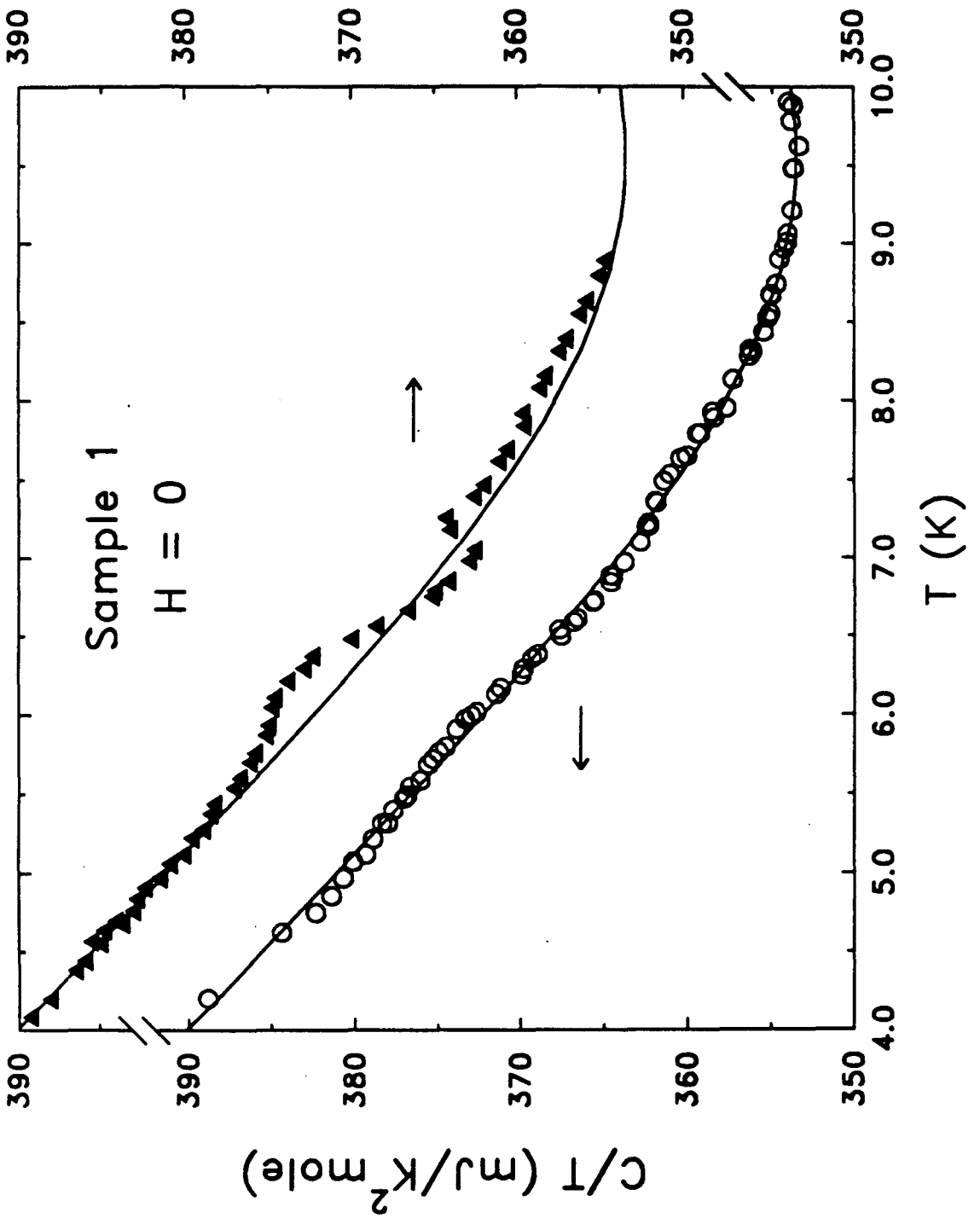

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FIG. 2

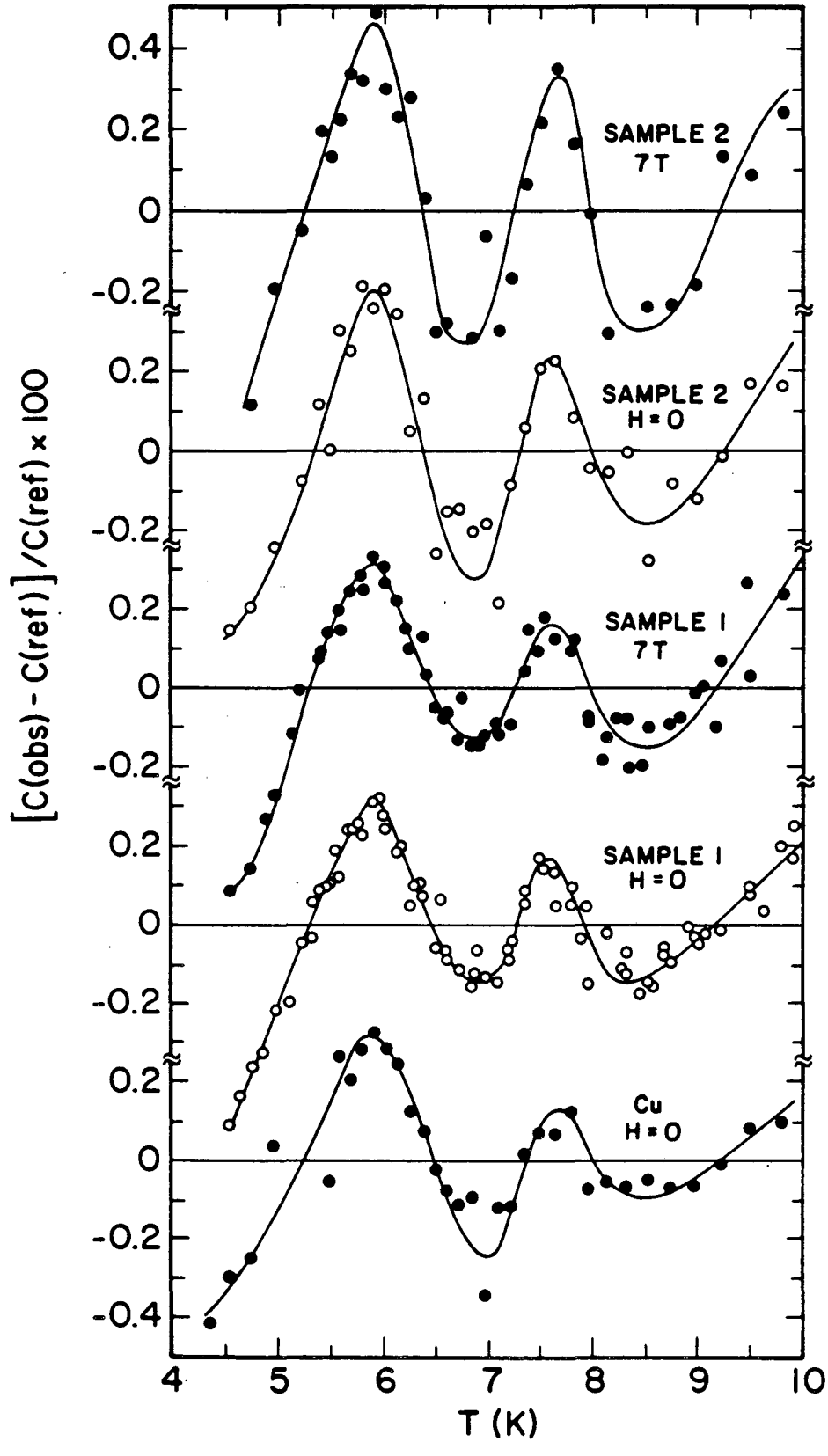

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FIG. 3

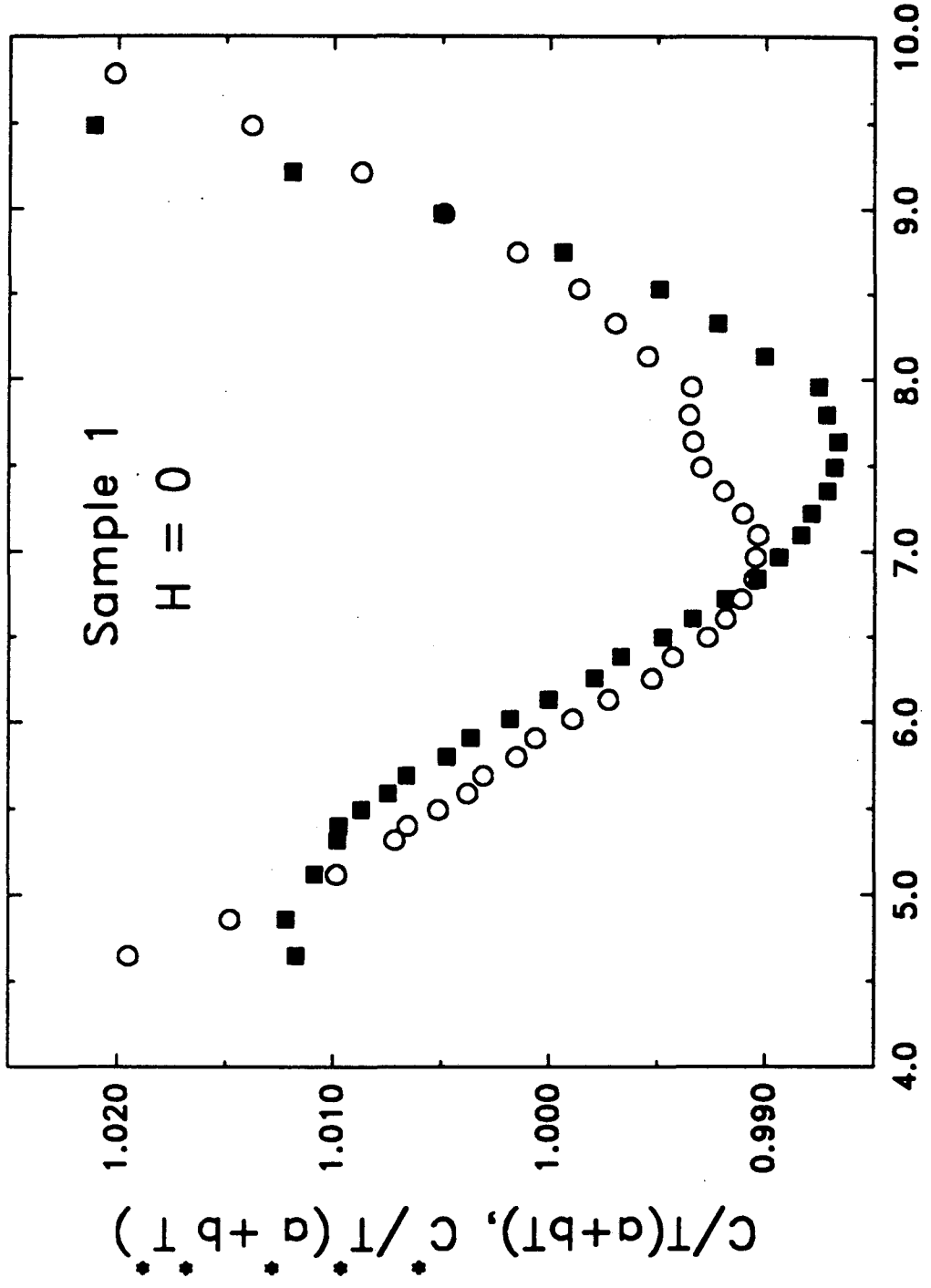

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FIG. 4

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