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Effect of Cu^{2+} Magnetic Moments on the Superconductivity of $YBa_2Cu_3O_7$: The Origin of the "Linear Term" in the Specific Heat; The Volume Fraction of Superconductivity

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ABSTRACT

The concentration (n_2) of Cu^{2+} magnetic moments located on the $YBa_2Cu_3O_7$ lattice is determined by the specific heat in a 7-T magnetic field. The discontinuity in specific heat at T_c [$\Delta C(T_c)$] and also the coefficient of the linear term [$\gamma(0)$] are correlated with n_2 in a way that suggests the operation of a pair-breaking mechanism that limits the transition to the superconducting states and provides a criterion for the volume fraction of superconductivity. $\gamma(0)$ can be represented by the sum of two contributions, both associated with Cu^{2+} moments, implying that there is no contribution that is an intrinsic property of the superconducting state. The interpretation of the data provides estimates of parameters relevant to the recognition of strong-coupling effects.

Running Title: Specific Heat of $YBa_2Cu_3O_7$

Key words: Specific heat, superconductivity

The Cu^{2+} moments in typical samples of $YBa_2Cu_3O_7$, YBCO, can be classified, at least approximately, as being of one of two different types. One type, present in concentration n_1, resides in impurity phases such as $BaCuO_2$ and orders at temperatures near 10K. The contribution of the specific heat, C, of $BaCuO_2$ associated with the ordering of these moments to the zero-field "linear term", $\gamma(0)T$, in the heat capacity of YBCO samples is well known [1]. The second type of Cu^{2+} moment is associated with the low-temperature, zero-field "upturn" in C/T that is observed in YBCO samples. That upturn is the high-temperature tail of a Schottky-like anomaly that has a maximum well below 1K [2] and which is broadened by a distribution of internal fields. In the presence of a sufficiently strong applied magnetic field, H, the anomaly is shifted to higher temperatures that are consistent with Cu^{2+} moments and the values of H [3]. In that case the anomaly is represented, within the experimental accuracy, by the Schottky expression if n_2 is sufficiently low; for higher values of n_2 the broadening of the anomaly by internal effective fields is still significant. These moments, with concentration n_2, affect the transition to the superconducting state (see below) and are therefore located, at least in substantial measure, on the YBCO lattice.

The components of C for a YBCO sample, represented by the solid curves in Fig. 1 for $T \leq 10K$ and for $H=0$ and 7T, are: the field-independent lattice specific heat, C_ℓ; the field-dependent linear term $\gamma(H)T$; the contribution associated with the ordering of the n_2 Cu^{2+} moments, $C_m(H)$; and a hyperfine specific heat that is present only in applied fields, $C_h(H)$. [$C_h(H)$ is clearly distinguished from the zero-field upturn in C/T, $C_m(0)$, by its dependence on H and T: in this temperature interval, $C_h(H)$ is within experimental error proportional to H^2/T^2, with a proportionality constant that can be understood in terms of the interaction of the nuclear magnetic moments with H; a fit to $C_m(0)$ requires a number of terms in inverse powers of T, reflecting both the higher characteristic temperature of the ordering of the electronic moments, and the broadening of the anomaly associated with the distribution of the Cu^{2+}-Cu^{2+} interactions.] The zero-field linear term, i.e., $\gamma(0)$, has received considerable attention, but there is no concensus as to its origin; its field-dependence, linear in H to within experimental accuracy, however, is consistent with the mixed-state electronic contribution that is well known in conventional superconductors. The solid curve labelled $C_m(7T)$ is a Schottky anomaly that determines the value of n_2. The data points in Fig. 1 are experimental data for 7T from which C_ℓ, $C_e(7T)$ and $C_h(7T)$ have been subtracted to give an indication of the reliability with which n_2 is determined.

In the following discussion of correlations between various parameters, data for 12 YBCO samples, including two Zn-doped samples, are used. For one of the Zn-doped samples n_2 is within the range of values covered by the undoped samples, and its properties are indistinguishable from those of the undoped samples. The other Zn-doped sample extends the range of values of n_2 by 15%. It is known from other work [4] that Zn doping produces Cu^{2+} moments.

The influence of the n_2 moments on the superconducting-state properties is shown most directly by the correlation of n_2 with the discontinuity in C at T_c, $\Delta C(T_c)$. Several methods of estimating $\Delta C(T_c)$ are represented in Fig. 2: The dashed lines represent a linear extrapolation of C from $T > T_c$ and $T < T_c$, and an entropy-conserving construction that gives $\Delta C(T_c)$. An extrapolation from $T > T_c$ based on a harmonic-lattice fit to high-temperature data [5], represented by the dash-dot curve would give essentially the same result, as does the fit with a Gaussian distribution of T_c's which is represented by the solid curve. Fits that take into account fluctuation contributions also give similar results for the mean-field $\Delta C(T_c)$.

For the samples considered here, T_c is essentially constant, and the correlation of $\Delta C(T_c)/T_c$ with n_2, represented in Fig. 3, is equivalent to that of $\Delta C(T_c)$ with n_2. In assigning the straight-line fit to the data, the point near $n_2=0.009$ was given zero weight because that sample showed a small Meissner effect, suggesting that the specific heat anomaly was too broad to permit a reasonable estimate of $\Delta C(T_c)$. One obvious interpretation of Fig. 3 is that $\Delta C(T_c)$ measures the volume fraction of superconductivity, f_s, and that the $n_2=0$ value of $\Delta C(T_c)/T_c$, 77 $\mathrm{mJ/mole \cdot K^2}$, is characteristic of an ideal, fully superconducting sample. This interpretation is consistent with other measures of f_s that can be derived from specific-heat data. For example, $d\gamma(H)/dH$ should also be proportional to f_s, and, as shown in Fig. 4, it is proportional to $\Delta C(T_c)$.

In addition to the correlation of $\Delta C(T_c)$ with n_2, there is an independent correlation of $\gamma(0)$ with n_2. The solid triangles in Fig. 1 represent $\gamma(0)$ as a function of n, where n is the total concentration of Cu^{2+} moments determined from high-temperature susceptibility measurements. The correlation of $\gamma(0)$ with n is similar to that reported by the Geneva group [6] -- the data scatter widely, but the relation is approximately linear. A fit with $\gamma(0)=\gamma_0+\gamma' n$ gives $\gamma_0=2.5$ $\mathrm{mJ/mole \cdot K^2}$ with an rms deviation of 34%. The concentration of Cu^{2+} moments in impurity phases can be calculated as $n_1=n-n_2$, and taking n_1 and n_2 as independent variables, $\gamma(0)=\gamma_0+\gamma_1 n_1+\gamma_2 n_2$, gives $\gamma_0=0.1\pm0.8$ $\mathrm{mJ/mole \cdot K^2}$, an rms deviation of 11%, and a value of γ_1 that is reasonable in relation to the properties of $BaCuO_2$. The n_1- and n_2-proportional components of that fit are represented by the open circles and open squares in Fig. 5. Eliminating the γ_0 term does not appreciably affect the fit. Thus, for these samples, and within the experimental uncertainty, there is no evidence for a contribution to $\gamma(0)$ that is an intrinsic property of the superconducting state. The Geneva group [7] has reached a similar conclusion by applying a different, but apparently related, criterion to the samples studied there.

The linear decrease in $\Delta C(T_c)$ and the linear increase in $\gamma(0)$ with increasing n_2 are strongly reminiscent of the pair-breaking action of magnetic moments in conventional superconductors. For conventional superconductors the operation of a pair-breaking mechanism and the associated occurrence of gapless superconductivity are characterized by linear relations between $\Delta C(T_c)$, n_2 and γ, but also by a linear decrease in T_c with increasing n_2 -- to $T_c=0$ for a value of n_2 comparable to that for which $\Delta C(T_c)=0$. The behavior of YBCO differs conspicuously from that of conventional superconductors in the n_2 dependence of T_c: for a value of n_2 for which $\Delta C(T_c)$ has decreased by more than a factor of 2, there is no significant change in T_c. These comparisons with pair breaking and gapless superconductivity in conventional superconductors, suggest a somewhat different model for YBCO, one that is also suggested by the very short coherence length, ξ: Superconductivity is suppressed by the pair-breaking interaction, but because ξ is small the result is a mixture of normal regions in the vicinity of magnetic moments, and superconducting regions with T_c unchanged elsewhere, rather than gapless superconductivity with T_c uniformly depresseed everywhere. The n_2 dependence of $\Delta C(T_c)$ is a measure of the volume fraction of superconductivity. The observed linear relation between $\Delta C(T_c)$ and n_2 is reasonable for this model for values of n_2 that are not too high, but it does not have the theoretical basis established for all n_2 for gapless superconductors. The Meissner effect, and other measures of the volume fraction of superconductivity derived from C data [8], which correlate with $\Delta C(T_c)$, are also consistent with the model.

With the use of the interpretation of specific-heat data outlined above, and in particular the values of f_s, it is possible to estimate the normal-state value of γ in several ways: (1) If $\gamma_2 n_2$ is extrapolated to the value of n_2 at which superconductivity disappears, i.e., at which $\Delta C(T_c)=0$, the value 16 mJ/mole$\cdot$K^2 is obtained. (2) The assumption that $\gamma(0)$ is linear in H in the mixed state, empirically valid as an approximation for conventional type-II superconductors, permits an estimate of γ by extrapolating $\gamma(0)$ to H_{c2}. For a polycrystalline sample $\overline{H_{c2}}=65$ T [9] and $\gamma=[d\gamma(H)/dH]\overline{H_{c2}}=18$ mJ/mole$\cdot$K^2. (3) Strong-coupling effects modify the temperature dependence of C_{es} generally and the shape of the anomaly at T_c in particular. These effects can be represented by the phenomenological "α" model [10] in which $2\Delta_0/k_BT_c$ is taken as an adjustable parameter. By fitting experimental data near T_c with this model both $2\Delta_0/k_BT_c$ and γ can be obtained. A preliminary analysis [8] gives $2\Delta_0/k_BT_c=7$ and $\gamma=14$ mJ/mole$\cdot$K^2. In the following, the average value, $\gamma=16\pm2$ mJ/mole$\cdot$K^2, is taken as a basis for comparison with band-structure calculations and for the evaluation of strong-coupling effects.

Massidda et al. [11] and Krakauer et al. [12] have obtained 16 and 13 mJ/mole$\cdot$K^2, respectively, for the bare or band-structure values, γ_{bs}. With $\gamma=16$ mJ/mole$\cdot$K^2, and $\gamma=\gamma_{bs}(1+\lambda)$, the corresponding values of the electron-phonon coupling constant, λ, are 0 and 0.2. Although $\lambda \leq 0.2$ does not correspond to strong phonon coupling, there is evidence of strong-coupling effects from the specific-heat data: The ratio $\Delta C(T_c)/\gamma T_c = 4.8$, which is to be compared with the weak-coupling BCS value, 1.43, is one example of an indication of extreme strong coupling. The value of $2\Delta_o/k_B T_c$, which is double that obtained in the weak-coupling limit is another. These indications of strong coupling together with the small value of λ are consistent with other evidence that the coupling is strong, but not entirely phonon-mediated.

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FIGURE CAPTIONS

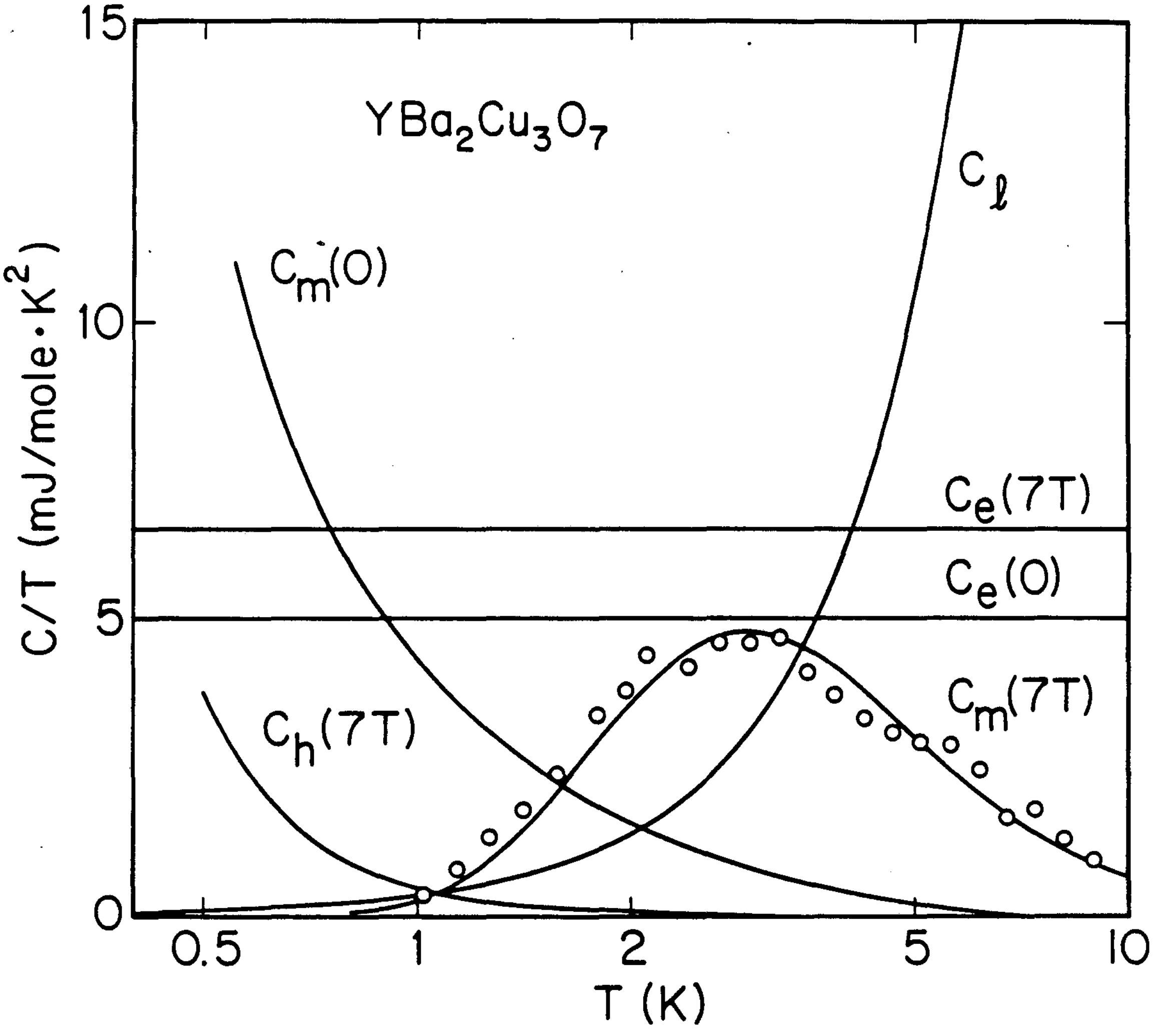
Figure 1. The components of the specific heat of a YBCO sample.

Figure 2. Analysis of the specific heat anomaly at T_c to obtain $\Delta C(T_c)$.

Figure 3. Correlation of $\Delta C(T_c)$ with n_2.

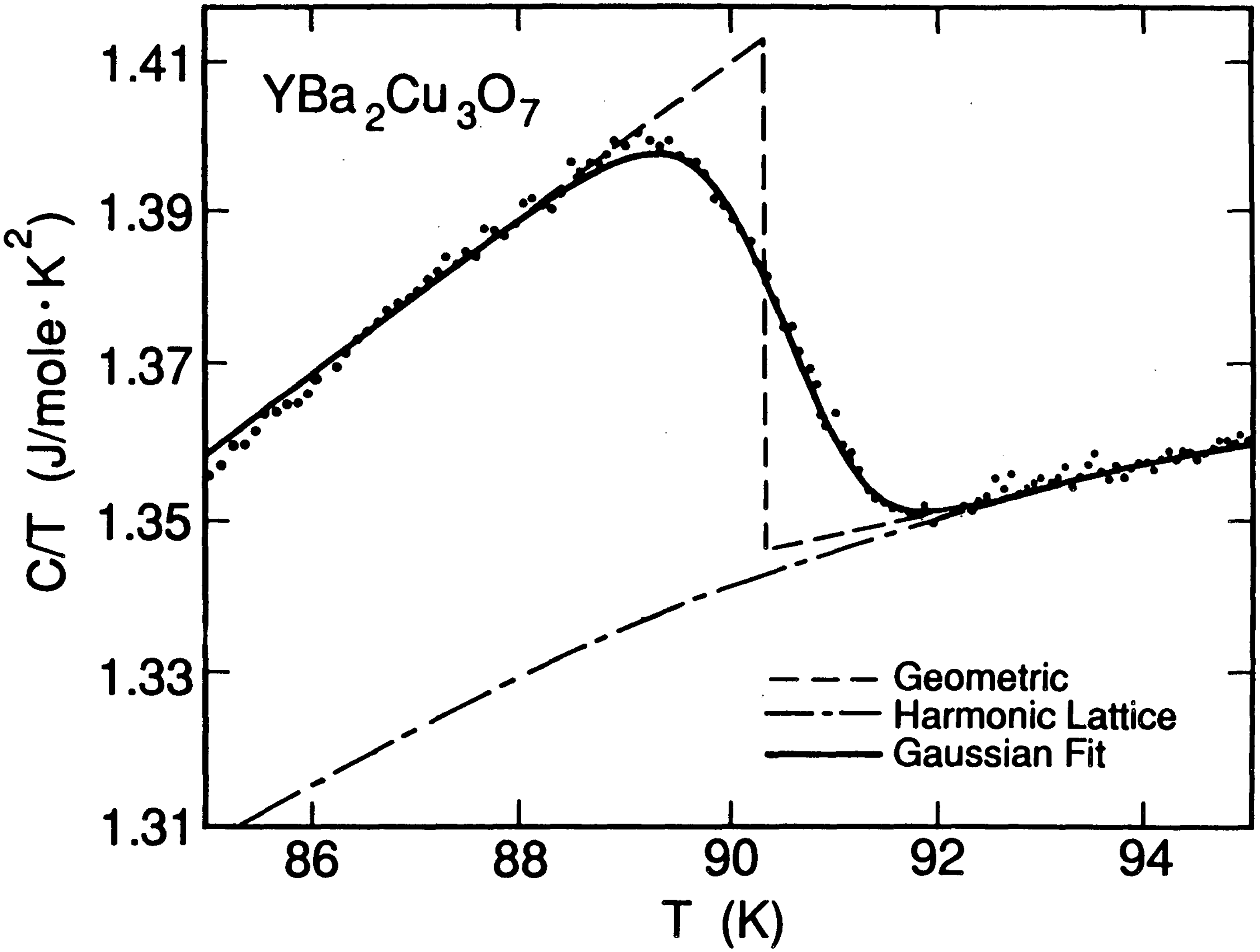
Figure 4. Correlation of $d\gamma(H)/dH$ with $\Delta C(T_c)$.

Figure 5. Correlation of $\gamma(0)$ with various concentrations of Cu^{2+} moments. See text for definitions.



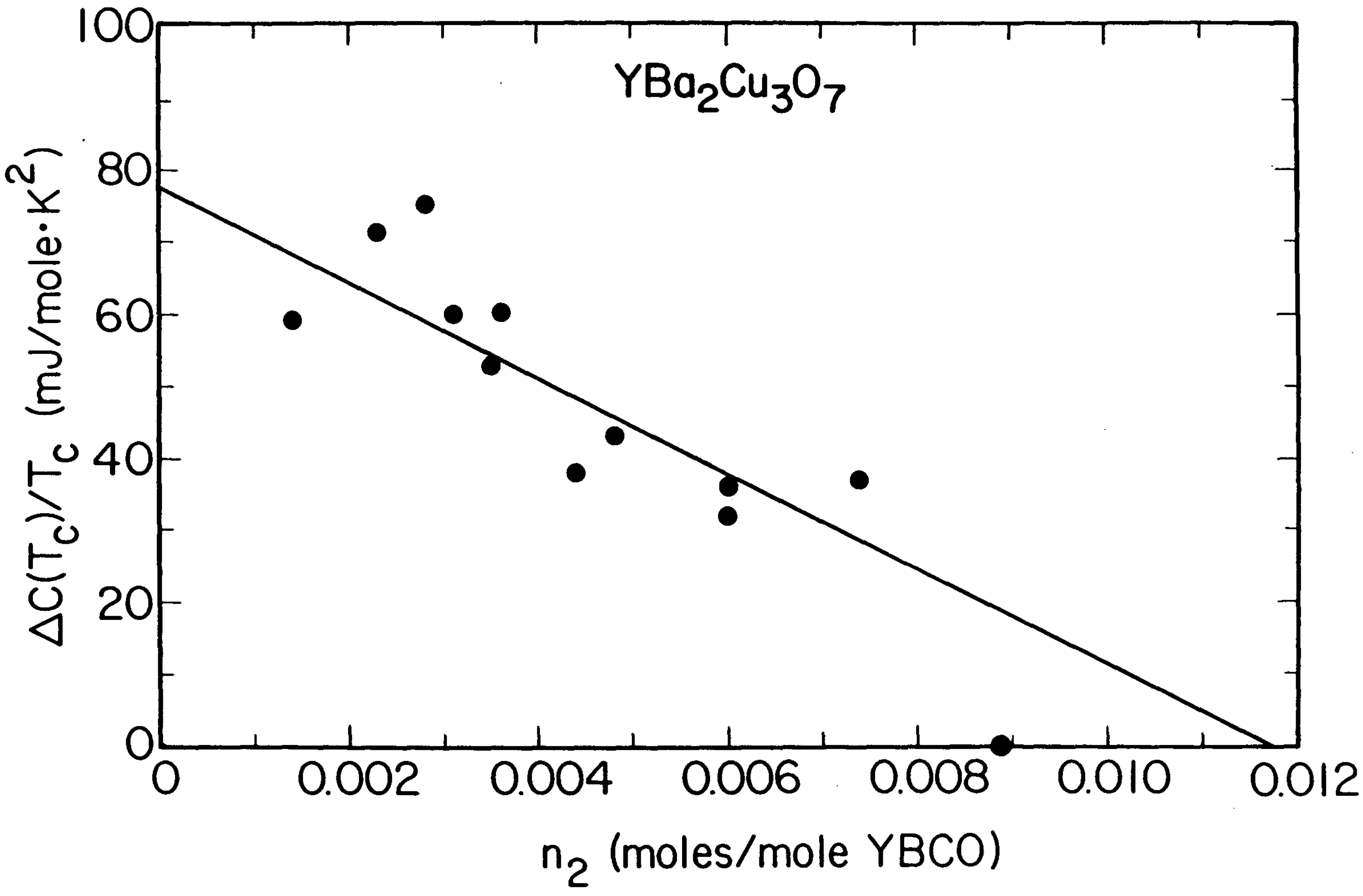
XBL 904-1224

FIG.1



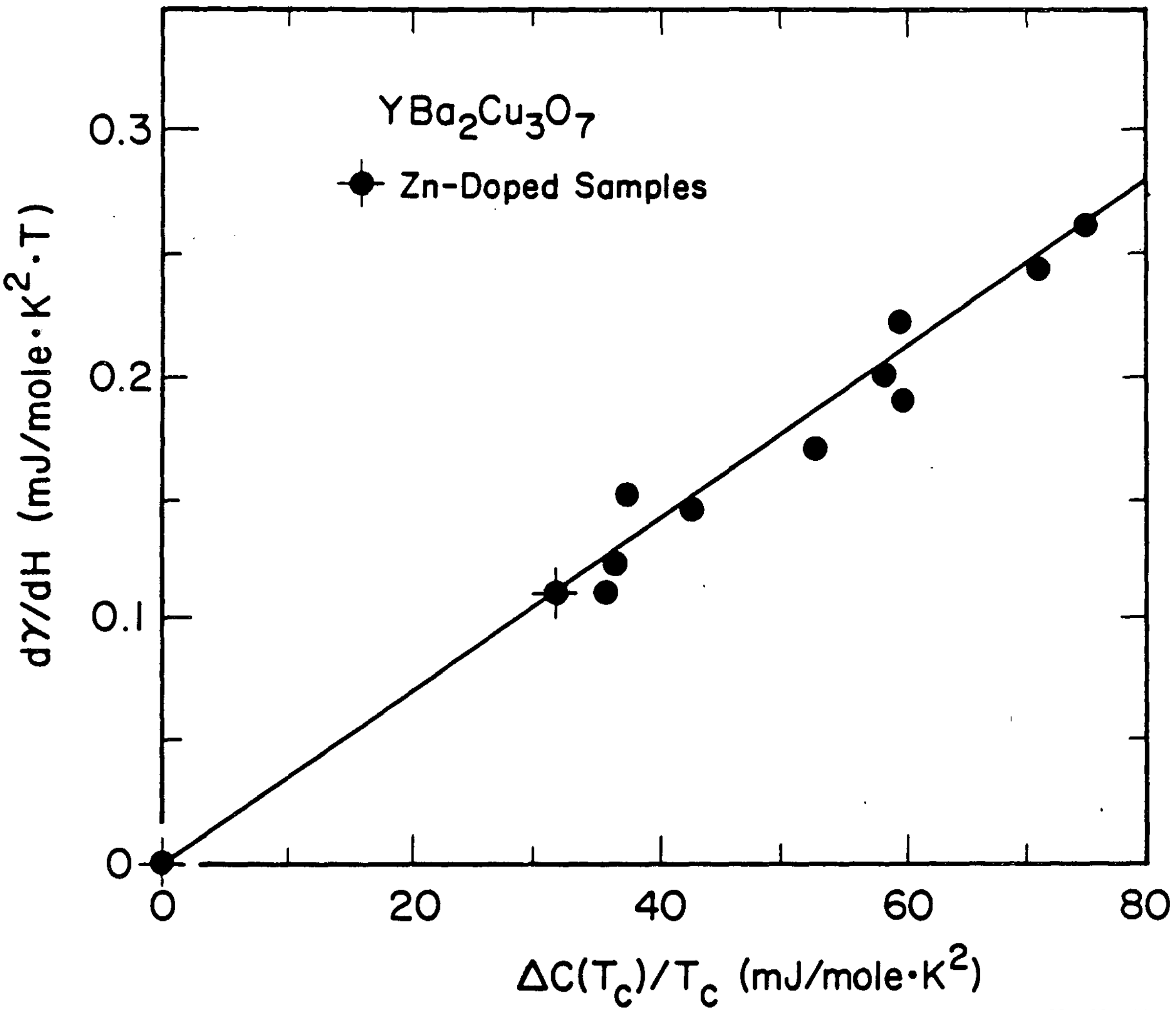
XBL 901-63

FIG.2



XBL 903-786A

FIG.3



XBL 902-444

FIG.4

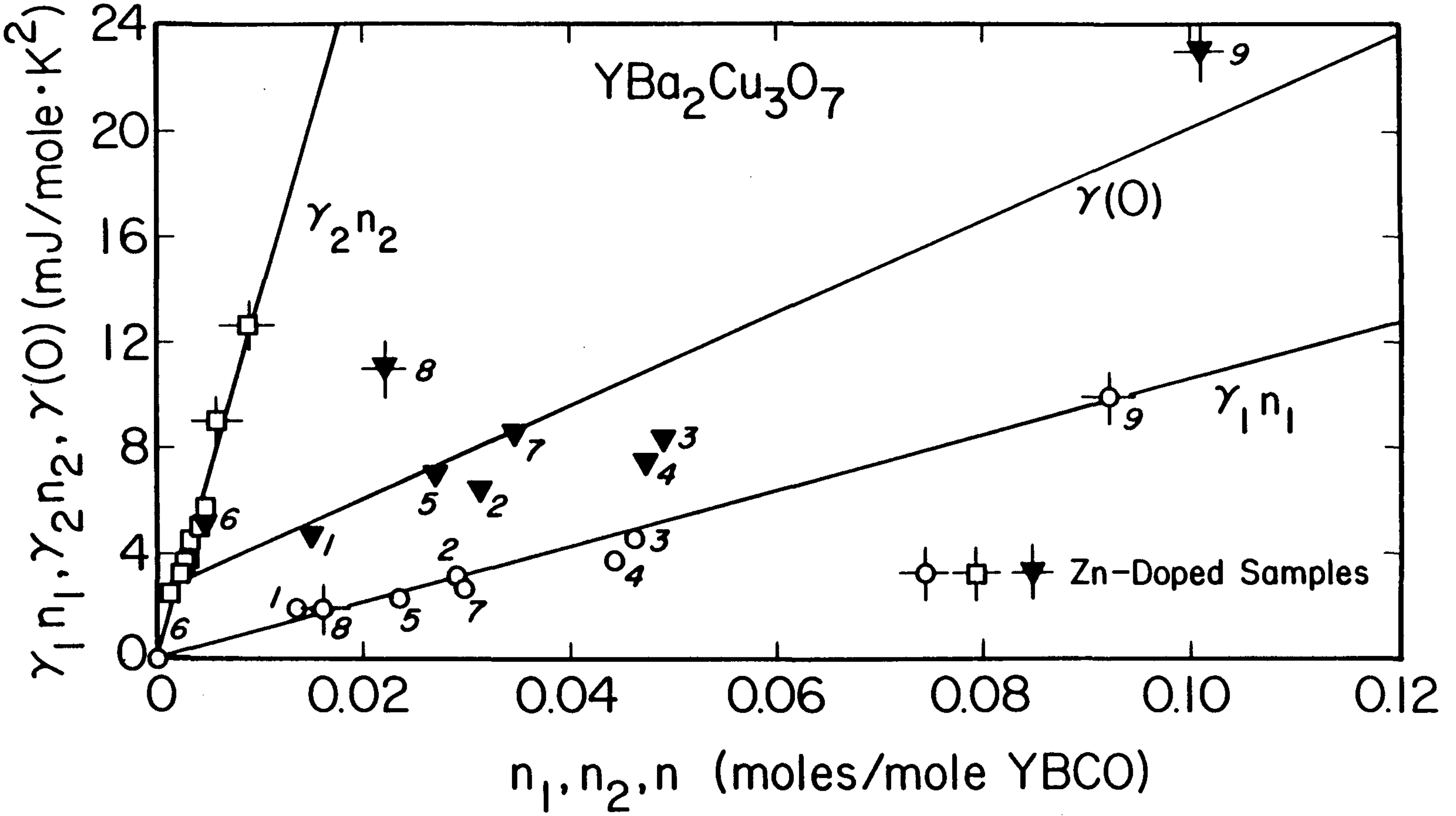


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FIG.5

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