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MAGNETIC ORDERING IN DILUTE $Gd_xEu_{1-x}Ba_2Cu_3O_{7-\delta}$ SUPERCONDUCTORS

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We have measured the specific heat of $Gd_xEu_{1-x}Ba_2Cu_3O_{7-\delta}$ ($0 \leq x \leq 1$). The data show a λ-type anomaly at high concentrations and a broad Schottky type anomaly at intermediate and low concentrations. We compare this behavior with theoretical predictions for a 2-dimensional Ising system.

The occurrence of long range antiferromagnetic ordering at low temperatures in the high-T_c superconductor $GdBa_2Cu_3O_{7-\delta}$ has been established through specific heat and magnetic susceptibility measurements (1-3) as well as neutron diffraction (4) and Mössbauer spectroscopy (5). Two important questions remain: what is the nature of the magnetic interactions, and what is the dimensionality of the magnetic ordering? Recent neutron diffraction experiments indicate the existence of a 3-d antiferromagnetic order (4). However, van der Berg et al. (6) have proposed a description based on a magnetic transition with 2-d Ising character, driven by superexchange interactions. This 2-d character is supported by the crystalline structure of the Gd lattice and the magnetic behavior above the Néel temperature, T_N. We report here specific heat data for $Gd_xEu_{1-x}Ba_2Cu_3O_{7-\delta}$ that also support this description.

The samples were prepared by sintering mixed powders of Eu_2O_3, Gd_2O_3, $BaCO_3$, and CuO in an oxygen atmosphere, as described in (3).

The specific heat was measured down to 0.45K in a semiadiabatic calorimeter (7) using a standard pulse technique. In Fig. 1 we show the magnetic specific heat, C_m, obtained after subtracting the specific heat of a sample with x = 0. The data for $GdBa_2Cu_3O_{7-\delta}$ shows a λ-type anomaly peaked at $T_N = 2.24K$, as we have previously reported (2) and which is in agreement with other reports (1,6). Above T_N, the integrated area under the curve is much larger for this system than for typical 3-d antiferromagnets. This feature indicates that short-range magnetic order is important well above T_N. A measure of this effect is the entropy fraction, $S(T_N)/R \ln 8$, where $R \ln 8$ is the entropy calculated for Gd^{3+} free ions ($4f^7$; $^8S_{7/2}$). In order to estimate the entropy, S(T), we have integrated our experimental data for $C_m(T)/T$ vs. T, extrapolating linearly to zero below 0.45K and assuming $C(T) \propto T^{-2}$ above 5K. For $S(T \to \infty)$ we obtained 96% of the theoretical value. Only about 60% of this amount is removed below T_N. This entropy fraction is much lower than expected (8) for 3-d systems (0.85-0.90) and supports the 2-d description of the magnetic ordering.

As $x \to 0$, the λ-type feature of C_m decreases in intensity and a broad Schottky type maximum develops, as shown in

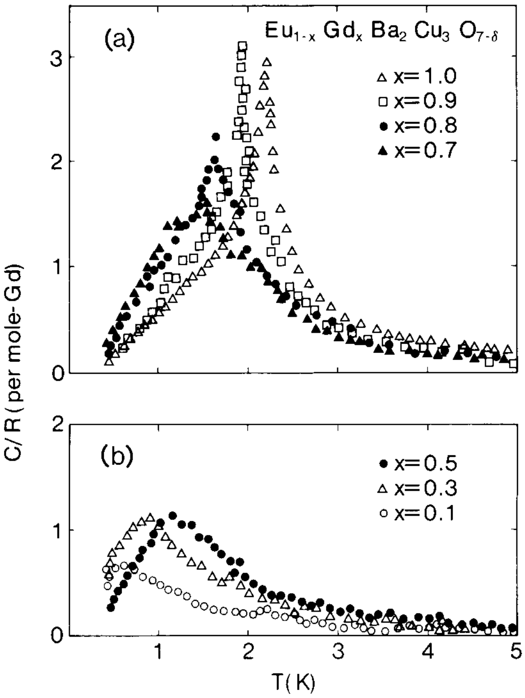

Fig. 1. Specific heat of $Gd_xEu_{1-x}Ba_2Cu_3O_{7-\delta}$ vs. temperature. Note (a) the λ anomaly for x > .7 and (b) the broad Schottky anomaly for x < .5.

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Fig. 1(b). This behavior has been predicted for dilute 2-d Ising lattices (9) and interpreted as a progressive freezing out of local disorder. In our case the λ-type maximum is observed for $0.8 \leq x \leq 1$ and the broad maximum appears for $x \leq 0.5$. For $x \sim 0.7$, it is possible to barely distinguish a second peak below the broad maximum. In Fig. 2 we present the temperature at the maximum C_m, T_{max} vs. x.

The λ-peak shifts to lower temperatures with initial normalized slope, $dt/dx \equiv d[(T_{max}(x)/T_{max}(x = 1)]/dx = 1.32$. This value is slightly smaller than the predictions of Monte Carlo calculations ($dt/dx = 1.47$) for the 2-d Ising system (10). The intercept of $T_{max} = 0$ occurs at a finite concentration, which suggests the existence of a percolation threshold. This would indicate that the ordering mechanism for samples with $x \sim 1$ arises from short-range interactions, probably due to superexchange through oxygen ions.

When only nearest-neighbor interactions between Gd ions are taken into account, the maximum of the broad anomaly has been predicted to occur (9) at temperatures of the order of J/k_B. For our samples this maximum is strongly concentration dependent, and it is therefore not possible to estimate a unique value for J/k_B. This may indicate the existence of a spread of magnetic coupling constants with more distant neighbors, characteristic of longer range interactions. We suggest that dipolar couplings between Gd moments ($E_{dd} \propto \mu^2/R^3$) may play a significant role at low concentrations in addition to superexchange interactions.

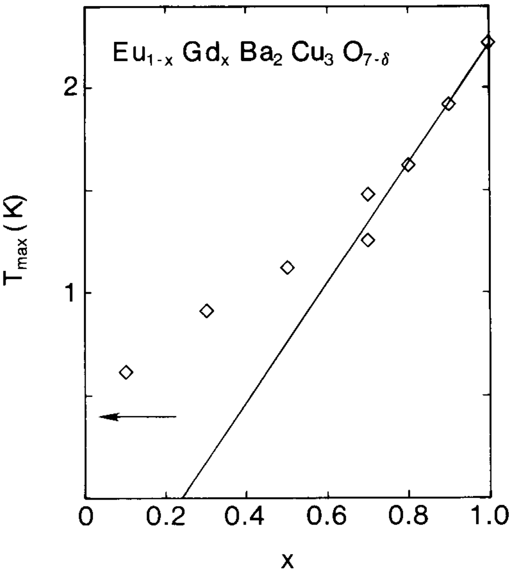

Fig. 2. Temperature, T_{max}, for which C_m is a maximum vs. x, the fractional concentration of Gd. The arrow indicates the calculated position of the crystal field Schottky anomaly.

The Schottky anomaly tail observed for $x = 0.02$ is related to the crystal field splitting, as discussed in (3). The single ion anisotropy energy associated with this interaction, $E_a = 21 \mid D \mid /2 \sim 1.4$K, is of the same order of magnitude as the calculated dipolar coupling energies (5) and it should be included when magnetic anisotropy effects are evaluated. If D were negative, the orientation of the magnetic moments would be favored along the c-axis, as found experimentally. As a result, the magnetic interaction between Gd moments might be described by an Ising Hamiltonian as proposed in (6).

In conclusion, we have shown that the specific heat data for the $Gd_xEu_{1-x}Ba_2Cu_3O_{7-\delta}$ dilute antiferromagnetic system presents characteristics of a 2-dimensional Ising model; i.e. a large tail above T_N, and a relatively large rate of depression of T_N upon dilution. The concentration dependence of T_N seems to indicate that, besides superexchange between nearest Gd atoms, magnetic interactions between farther neighbors should be considered.

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