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Reentry-Like Features in $YBa_2Cu_3O_7$ and $GdBa_2Cu_3O_7$

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Abstract

The diamagnetic field-cooled susceptibility of $YBa_2Cu_3O_7$ and $GdBa_2Cu_3O_7$ curves upward at low temperature and crosses over to a positive value at temperature T^* which increases with increasing field. We demonstrate that the low-temperature phase is superconducting, and the reentry-like features are a result of a paramagnetic background inherent in this class of materials. The relevance of this behavior to the pairing mechamism is discussed.

Since Bednorz and Müller [1] reported on the possibility of a high temperature superconductor, the highest transition temperature T_c has advanced substantially [2–4]. Wu *et al.* [2] were the first to report on a Y–Ba–Cu–O system with $T_c \approx 90$ K. Several compounds of similar composition are known today to have $T_c \geqslant 90$ K [5]. In this work we present magnetic measurements for two super-conductors of the 90 K family, $YBa_2Cu_3O_7$ and $GdBa_2Cu_3O_7$, and focus attention on field-induced reentry-like features at low temperature. Most of the results for $YBa_2Cu_3O_7$ have already been reported in Ref. [6] and they are reported briefly here for the sake of comparison with the results for $GdBa_2Cu_3O_7$.

The Y sample was prepared from a mixture of $BaCo_3$, Y_2O_3 and CuO powders (at least 99.9% pure) in stoichiometric proportion according to the formula $YBa_2Cu_3O_7$. Gd_2O_3 replaced Y_2O_3 in preparing the Gd sample. Finely ground powders were pressed into a pellet approximately 1.5 cm in diameter, and heated to 900°C for 16 h in flowing oxygen. The product was then quenched to room temperature, reground and heated again to 900°C for 48 h, then cooled to ambient temperature.

The magnetic measurements were carried out on a superconducting quantum interference device (SQUID) magnetometer. The sample is zero-field-cooled (ZFC) to low temperature where a field H (20 Oe $\leqslant H \leqslant$ 10 kOe) is applied and the ZFC ("shielding") branch of the magnetization is measured while temperature is increased. The sample is then cooled in the presence of the field and the field-cooled (Meissner) branch of the susceptibility is measured while temperature is increased. A typical ZFC/FC run for $GdBa_2Cu_3O_7$ is presented in Fig. 1. The striking similarity between the behavior exhibited in Fig. 1 and the magnetic behavior of spin glasses has already been pointed out for other high-T_c superconductors [6, 7] and will be discussed elsewhere. Here we emphasize another feature which characterizes the new high-T_c superconductors: the upward turn of the magnetization at low temperature, which can be clearly observed in Fig. 1. Figures 2 and 3 exhibit typical field-cooled (FC) runs for various fields for $YBa_2Cu_3O_7$ and for $GdBa_2Cu_3O_7$. For both samples below a field-dependent temperature $T^*(H)$ the measured magnetization is *not* diamagnetic.

Does T^* signal a reentry temperature? This is probably not the case. We demonstrate that the low-temperature phase is still superconducting by measuring the trapped flux which characterizes the mixed phase in type II superconductors. This is done by turning the field off at low temperature and measuring the remanent magnetization of the FC branch. We find that the remanent magnetization, which reflects the trapped flux, always has an initial positive value *larger* than the magnetization in the presence of the field. *This implies that the induced magnetization still has a substantial diamagnetic component even though the measured low-temperature signal is positive.* The remanent magnetization decays slowly towards a stable state, exhibiting another aspect of the glassy nature of these systems [6–9]. The nature of the time dependence is discussed elsewhere [10].

What is the origin of the low-temperature positive contribution? In order to answer this question we first analyze the high-temperature paramagnetic behavior. We find that well above T_c the paramagnetic susceptibility χ_{PM} is composed of

$$\chi = \chi_0 + C/(T - \Theta) \qquad (1)$$

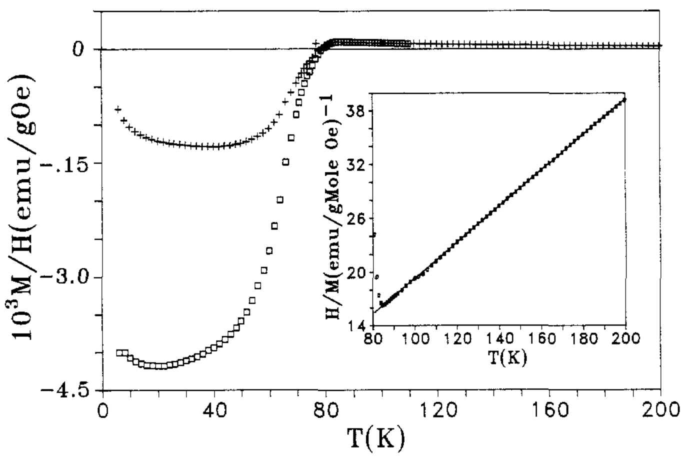

Fig. 1. Zero-field-cooled and field-cooled branches of M/H of $GdBa_2Cu_3O_7$ measured in H = 0.5 kOe. Inset: Inverse susceptibility H/M above T_c. The solid line is the paramagnetic susceptibility, eq. (1), fitted for data points between 110 K and 200 K.

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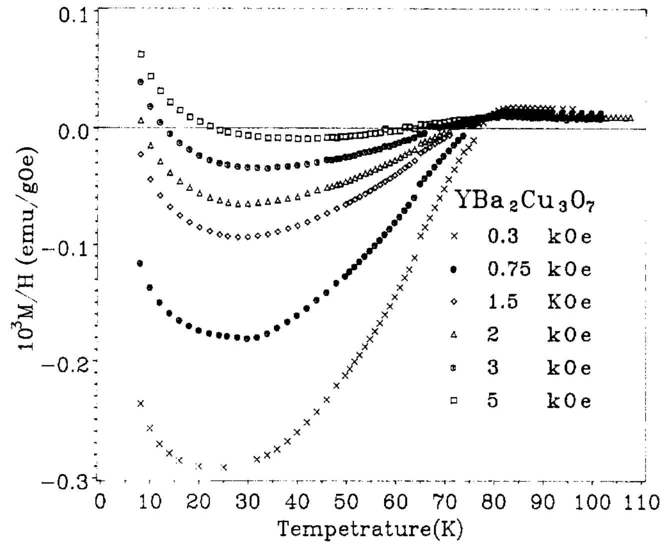

Fig. 2. M/H field-cooled branches of $YBa_2Cu_3O_7$ measured in the indicated fields (from Ref. [6])

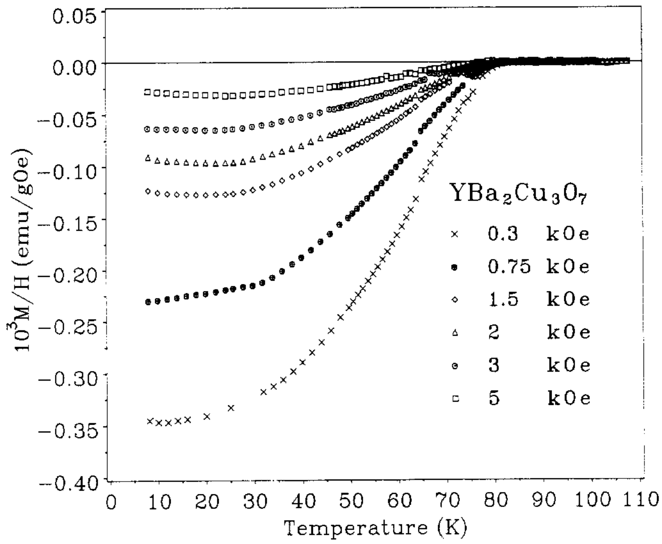

Fig. 4. Diamagnetic contribution ($M/H - \chi_{PM}$) for the field-cooled data of Fig. 2, (from Ref. [6]).

where χ_0 is a Pauli temperature-independent susceptibility. We find quite surprisingly high values for χ_0: 3.4×10^{-4} emu/mole and 1.9×10^{-4} emu/mole the Y and Gd samples respectively. These values are larger by more than an order of magnitude than the usual metallic Pauli susceptibility. Similar high values have been found in the La superconductors ($T_c \approx 35$ K) and were interpreted to indicate strong electron-phonon interaction [11]. The Curie constants C which are found by fitting the high-temperature data to eq. (1) are 0.5 and 4.9 Kemu/mole, for the Y and Gd systems respectively. This implies a localized moment contribution of approximately $2\,\mu_B$ and $6.7\,\mu_B$ per unit cell for the Y and Gd system respectively. The values obtained for $YBa_2Cu_3O_7$ are due to Cu ions whereas the higher value for the Gd system is due mainly to the contribution from Gd moments. The values of the Curie–Weiss temperature were found to be 0 K and 4 K for the Y and Gd samples respectively. We emphasize that the results quoted here are sample dependent and they might be scattered significantly from sample to sample. However, the qualitative arguments of this article are valid in all cases.

We demonstrated above that the low-temperature magnetization is composed of paramagnetic and diamagnetic contributions. We assert that the low-temperature paramagnetic contribution is of the same origin as high temperature ($T > T_c$) susceptibility. To test this assertion we extrapolate the paramagnetic susceptibility, eq. (1) to low temperatures, using the parameters described above. We then substract the extrapolated χ_{PM} from the raw data. The "pure" diamagnetic contribution χ_d obtained by such a procedure is displayed in Fig. 4 for the data of Fig. 2. It is apparent that χ_d is indeed diamagnetic at all temperatures below $T_c(H)$ and it behaves as expected for the diamagnetic susceptibility for superconductors except for a slight upward curvature which is still observed at low temperatures. This probably indicates that θ is not exactly zero for the Y system (we estimate the error to be of order 1 K), and thus the paramagnetic contribution at very low temperatures are more enhanced than assumed here. Similarly, χ_d for the Gd system is indeed diamagnetic at all temperatures below $T_c(H)$ but in this case it curves downward at low temperature (Fig. 5). This downward curvature is a result of the assumed divergence of χ_{PM} at 4 K. Actually it is known that $GdBa_2Gu_3O_7$ exhibits an antiferromagnetic transition at 2 K [12, 13] and this sheds doubt, of course, on the validity of the extrapolation at this vicinity.

Are the magnetic features which have been described above relevant to the understanding of high-temperature superconductivity? Strong electron–phonon couplings to O-breathing mode vibrations have been suggested [14] to

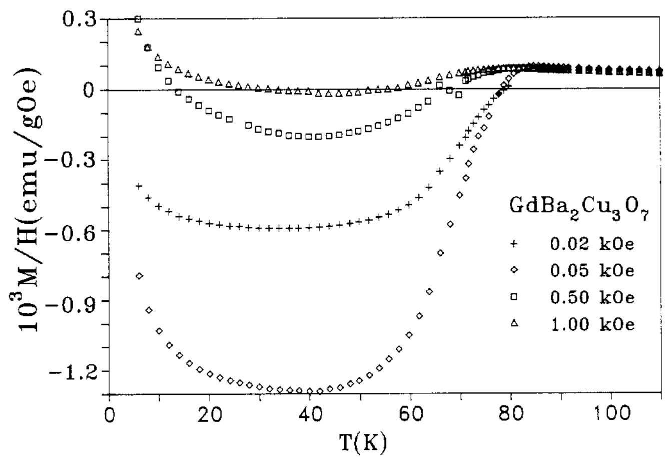

Fig. 3. M/H field-cooled branches of $GdBa_2Cu_3O_7$ measured int he fields indicated.

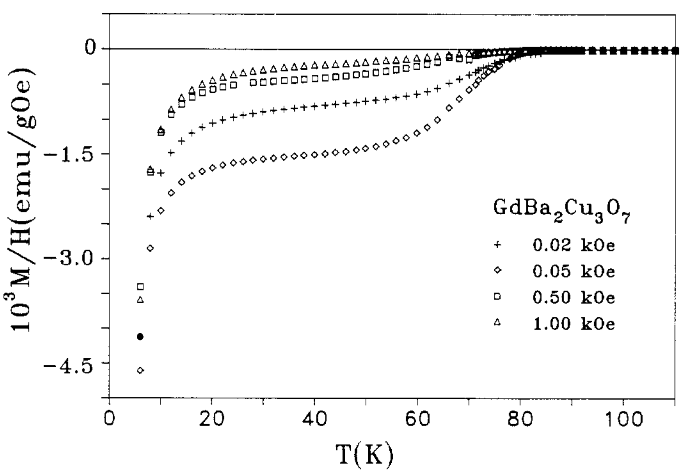

Fig. 5. Diamagnetic contribution ($M/H - \chi_{PM}$) for the field-cooled data of Fig. 3.

explain the high T_c values in the framework of the conventional BCS theory [15]. However, the lack of an isotope effect [16–18] casts serious doubts on the relevance of the electron-phonon mechanism to high-T_c superconductivity. Non-phonon mechanisms which are not related to ionic mass have been proposed. We refer, in particular, to a recent theory [9] which is related to the latter family of models and which predicts explicitly reentry-like features due to magnetic fluctuations. This model is based on antiferromagnetic (AFM) interactions [19] which give rise to pairing and high T_c and at the same time yields at low temperatures, strong paramagnetic contributions. According to this model, spin flip of two-neighbor electrons in a diluted AFM system is equivalent to an attractive pairing interaction similar to that described by bi-polarons. At high temperatures, below T_c, most of the thermal energy if found in the form of kinetic energy for electron pairs. At low temperatures, on the other hand, most of the energy goes towards spin flip which contributes a strong Curie-like paramagnetism in agreement with the present experimental data. Further experimental and theoretical work is undoubtedly needed to pursue this idea.

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