PHYSICAL REVIEW B VOLUME 43, NUMBER 1 1 JANUARY 1991

Magnetic rare-earth contributions to muon relaxation rates and frequency shifts in $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-\delta}$

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(Received 24 July 1989; revised manuscript received 11 June 1990)

Contributions of rare-earth (R) magnetic moments to muon frequency shifts and relaxation rates are obtained for superconducting Gd- and Er-based barium cuprates by comparison with a (nonmagnetic) Eu-based sample. The R-related contributions approximately scale with the paramagnetism. A model of R-spin-flip-induced local supercurrents is introduced, which is consistent with very short coherence lengths and qualitatively reproduces the enhanced frequency shifts and reduced R-related relaxation terms observed below T_c.

Within weeks after the discovery of the 90-K class of oxide superconductors,[1,2] experimental evidence appeared that demonstrated that the substitution of most magnetic rare-earth (R) ions into the yttrium site of $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_7$ had no significant effect on the superconductivity.[3-5] The decoupling of the rare-earth magnetism and superconductivity in the majority of these compounds is now a well-established fact, even though the R-Y site is sandwiched between the superconducting CuO_2 planes of the orthorhombic crystal structure. A detailed model of the superconducting interactions which explains this insensitivity to magnetic ions does not yet exist. However, we conjecture that in response to spin flips at the R sites, supercurrents must be induced in the immediate vicinity, which would then shield more distant parts of the structure from the spin-flip processes. Since the coherence length is very short in these materials, such shielding currents could be quite effective in isolating the R moments.

In this paper we report on muon-spin-relaxation (μSR) studies carried out at the Clinton P. Anderson Meson Physics Facility (LAMPF), in which the rare-earth contributions to the muon spin depolarization and frequency shifts were examined for $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_7$ superconductors with magnetic R ions Gd and Er. These data show the effects which we attribute to local shielding currents through two features: (1) reduction of the R-ion contribution to the depolarization in the superconducting state, and (2) a temperature-dependent diamagnetic frequency shift which is qualitatively different from the Meissner shift in samples based on nonmagnetic R ions, and considerably larger than expected from dipolar fields or sample demagnetization.

In μSR experiments, spin-polarized muons are implanted into a sample, where they probe the local magnetic environment at specific interstitial crystallographic sites. In the case of the $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_7$ materials, the most likely muon sites are μ-O bond sites between the CuO chains and the BaO planes.[6] The average internal magnetic field at the μ site and the variations in the field are obtained from the observed μSR frequency and the functional form for the relaxation of the muon polarization.

The data discussed here are for one Gd- and two Er-based $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_7$ polycrystalline samples with T_c above 90 K, and one sample of $\mathrm{GdBa}_2\mathrm{Cu}_3\mathrm{O}_{6.8}$ with $T_c = 81$ K. The μSR frequencies and Gaussian relaxation rates were obtained as a function of temperature while cooling in a 100-mT applied field. The basic data analysis and resulting penetration depths were previously published for the Gd samples[7] and a (nonmagnetic) Eu-based sample.[8] The data for the Er and Eu samples are compared in Fig. 1. In this paper we concentrate on the differences between the magnetic R-based systems and the nonmagnetic Eu-based sample.

The only significant differences in the μSR data between the magnetic and nonmagnetic R-based barium cuprates arise from the presence of R-ion magnetic moments. We argue that the Meissner shift is approximately the same for both cases and has the temperature dependence displayed by the Eu sample; and further, that a significant portion of the additional term in the shift for the magnetic case may be due to a supercurrent shielding response to R spin flips.

Typical temperature dependences of the μSR frequency and relaxation rates for magnetic-R-based $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_7$ are as shown for the Er-based sample in Fig. 1. The data for the Eu-based sample display the features due to superconductivity alone. The contributions from the R moments are the source of the differences between the two samples, which are shown as solid lines.

We chose to fit the Meissner-shifted μSR signal with a single Gaussian relaxation function. The difference between rate constants for the magnetic and nonmagnetic R-based samples shows a $1/T$ dependence. In fitting the

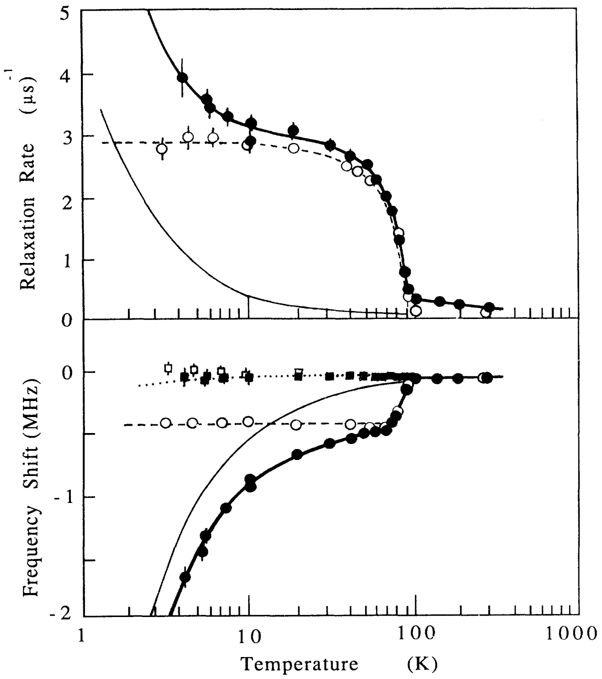

FIG. 1. Temperature dependence of (a) Gaussian μSR relaxation rates, and (b) μSR frequency shifts for the superconducting phase of $R\mathrm{Ba_2Cu_3O_{7-\delta}}$ with magnetic and nonmagnetic R ions. The solid line represents the R-moment contributions. Data: solid symbols—$R=\mathrm{Er}$ (magnetic); open symbols—$R=\mathrm{Eu}$ (nonmagnetic); circles—superconducting signal; squares—"normal" signal.

relaxation temperature dependence, three contributions to the rate constants were added in quadrature: a constant representing the effects of nuclear moments; a $1/T$ term due to the R moments; and below T_c, a term varying as $[1-(T/T_c)^4]$ from the two-fluid model of effects of magnetic-field penetration in the type-II mixed state. Results from these fits show significantly reduced magnitudes for the $1/T$ terms in the superconducting state, as compared to above T_c.

The origin of the $1/T$ terms is often argued to be the Korringa process, i.e., scattering of electrons from the R magnetic moment, which produces a fluctuation in the local fields sensed by the muon as an increase in field inhomogeneity. Within the single Gaussian analysis we cannot distinguish between a Korringa origin and any other source of field variation related to R paramagnetism. Our analysis shows that for $T>T_c$ the R-related term scales with the paramagnetism within experimental errors; i.e., as $\mu_R^2 H/T$, where μ_R is the R magnetic moment. Below T_c, the scaling is approximately as μ_R^2/T with a nonlinear field dependence as might be expected from the superconducting response to applied fields. The reduction of the R-related relaxation term below T_c ranges from a factor of $\sim 3-5$, implying a sample-dependent limitation on the reduced effectiveness of the R spin flips as a muon depolarization source in the superconducting state.

In evaluating the temperature dependence of the R-related frequency shifts Δf_R, some assumptions regarding the Meissner shift Δf_M are required. Rather than assume an R-related term proportional to $1/T$ and a constant Meissner shift, we use the knee observed in the data near 65 K (see Fig. 1) to experimentally define a value for Δf_M. Below 65 K the frequency in the nonmagnetic Eu sample remains constant, thus we take $f(65\ \mathrm{K})$ as the low-temperature Meissner-shifted value and define $\Delta f_R = f(T) - f(65\ \mathrm{K})$ as the contribution from the R ions. When defined in this manner and displayed as in Fig. 2, Δf_R clearly shows a $1/T$ temperature dependence as anticipated. Further, the magnitudes of Δf_R scale with μ_R^2 for the 90-K samples at 100 mT.

One source for an R-related frequency shift is demagnetization due to sample shape. Assuming a solid sample of the dimensions of our sintered disks, the calculated shift from this source is somewhat less than half the observed value. Since the samples are only 75% theoretical density, this may not be the appropriate treatment. An experimental estimate based on the largest shifts observed above T_c yields an extreme upper limit for demagnetization of less than 20% of the shifts seen in the superconducting state. A second estimate could come from the frequencies observed in an Er-based sample which was annealed to reduce the oxygen content to $\delta=0.8$. At the same fields, this sample shows a paramagnetic shift about 20% as large as the diamagnetic shift in the same disk when fully oxygenated. This shift should be related to the Er paramagnetism; however, Cu antiferromagnetism or "loose" Cu moments might cause complications. These differences, however, argue against demagnetization as the dominant factor, since effects due to the Er moments should be the same in the two cases.

We also observe a μSR signal which is very near the normal-state frequency and amounts to 12–15 % of the total intensity. The acceptance geometry of counters placed inside our cryostat precludes any significant signal

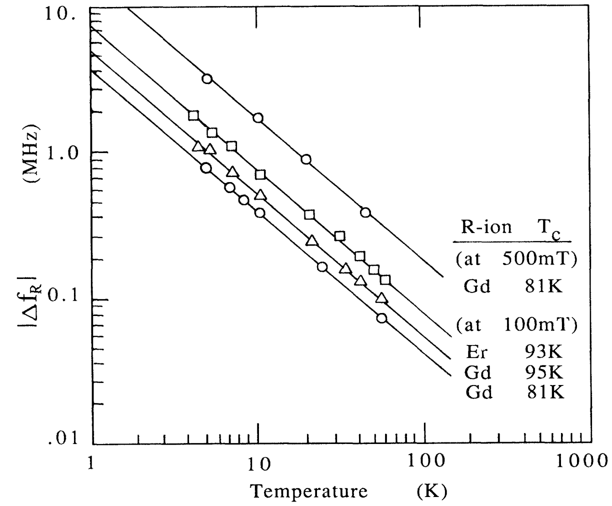

FIG. 2. R-related μSR frequency shifts for $R\mathrm{Ba_2Cu_3O_{7-\delta}}$ below T_c; display of the $1/T$ dependence.

arising from nonsample sources. We have experimentally confirmed that no detectable (nonsample) background signal is present. This weak second signal is represented by the squares and dotted line in Fig. 1(b). The frequency shows a small diamagnetic shift at low temperatures in the magnetic R cases and a very small paramagnetic shift for the Eu-based sample. Since all of these samples are single phase as determined by x-ray characterization (1–3 % sensitivity), we conclude that this signal is from twin and grain boundaries, or other regions which are (a,b)-axis disordered on a very short length scale and thus not superconducting. This weak signal should, therefore, give the shift for the normal state, including both dipolar and demagnetization effects.

In modeling the response of $R\mathrm{Ba_2Cu_3O_7}$ superconductors to rare-earth moments, we start from an extreme assumption of very high two-dimensional localization of the supercurrents into the $\mathrm{CuO_2}$ planes on either side of the R site. The lack of magnetic pair breaking from R ions suggests that the superconducting wave functions may not overlap with the R site. Recent measurements[9] yielding coherence lengths of ~ 1.5 Å in the c direction support this extreme 2D picture. The physical extent of fields from a single R moment is much shorter than the penetration depth for externally applied dc fields. Additionally, in a continuum model, the currents induced in a perfect conductor by a magnetic transition of a point dipole are highly localized. Therefore, as a first step, we have modeled the spin-flip-induced currents in the limit of extreme localization. In this model, when an R moment flips, the locally induced currents will oppose the change in magnetic flux between the nearest superconducting planes. Since the most likely muon site is located further from the R site than the nearest $\mathrm{CuO_2}$ planes, the muon should see reduced fluctuations from the R spin flips. The observed reduction in the rare-earth contribution to the μSR relaxation in the superconducting state is qualitatively consistent with this picture.

The rather large μSR frequency shift attributable to rare-earth sources provides a means of making some rough quantitative comparison with experimental numbers. In selecting a specific model for calculations we choose to place the magnetic field in the (a,b) plane because, far from the center of a fluxoid, the anisotropic magnetic penetration depth will rotate the field toward that plane.[10] Given likely muon sites, dipole fields of an (a,b)-oriented R moment will give a diamagnetic frequency shift, while a c orientation would yield a paramagnetic shift. These anisotropic effects imply that a diamagnetic shift is expected due to the mean local dipole fields from the R ions. However, the observed shift is more than an order of magnitude greater than predicted from this source.

Rather than rely solely on calculated demagnetization and dipolar shifts, a comparison is made with the diamagnetic shift in the weak "normal-state" μSR signal which, we argue, arises from material with the same relative positions of the R and muon sites as in the superconducting regions. These normal-state shifts are barely detectable at 100 mT, but are clear at 500 mT. Our best experimental estimate is that the rare-earth-related superconducting shift is 30±3 times larger than the "normal" shift.

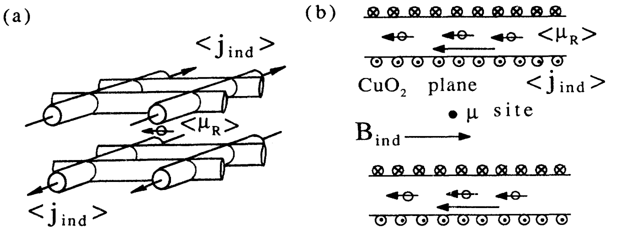

FIG. 3. Schematic representation of time-averaged local R-spin-flip-induced supercurrents as used in model calculations: (a) discrete currents, and (b) continuous current distribution in the $\mathrm{CuO_2}$ planes.

Two different approximations, as represented in Fig. 3, were used to make quantitative estimates of the expected spin-flip-induced frequency shifts. The first method [Fig. 3(a)], similar to a model used to explain magnetostriction data[11] in $\mathrm{GdBa_2Cu_3O_7}$, simulates the currents as an intersecting network of tubes about 1 Å in diameter representing the network of Cu-O wave functions. The second method [Fig. 3(b)] uses a set of 2D sheets of oppositely directed uniform current density representing the time-averaged induced currents in the two nearest planes, and results in oppositely directed uniform fields in the regions between planes. To estimate induced current magnitudes, we match the net magnetization and current-induced flux through the unit cell walls between the $\mathrm{CuO_2}$ planes for the region containing the R ions. Both methods yield a diamagnetic supercurrent-induced μSR frequency shift with an enhancement factor of about 50 over dipole fields, giving an upper limit for comparison with the experiment. Within the assumptions of our simplified model, these calculations imply maximum locally induced currents of about 10^{-4} A in the nearest current tubes for full cancellation of a single R spin flip. This translates to a current density of $\gtrsim 10^{10}\ \mathrm{A/cm^2}$ if carried over to bulk supercurrents. Additionally, upper and lower limits are implied for the persistence time of the induced currents. Given the experimental conditions, we estimate this range to be 10^{-3}–10^{3} s.

In conclusion, we have examined the contributions of rare-earth moments to the μSR frequency shifts and relaxation rates for $R\mathrm{Ba_2Cu_3O_7}$. These contributions scale with the rare-earth paramagnetism with a significant reduction in the R-related muon spin depolarization in the superconducting state, and yield an enhanced diamagnetic frequency shift. These effects must be properly removed when extracting such measurements as penetration depth from μSR data on magnetic rare-earth-based oxides. We attribute a significant fraction of these effects to R spin-flip-induced supercurrents. Such local shielding currents should be most effective for short c-axis coherence lengths as observed for these materials. Given the relative success of the simple model we employed, development of a detailed description of the spin-flip-induced response under less extreme assumptions should result in an increased understanding of the interactions between the decoupled rare-earth magnetism and superconductivity in the $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_7$ materials.

Portions of this work were supported by The Robert A. Welch Foundation, The Research Corporation, and The San Jose State University Foundation.

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