PHYSICAL REVIEW B VOLUME 49, NUMBER 17 1 MAY 1994-I

Muon-spin-rotation study of vortex states in $R\mathrm{Ba_2Cu_3O_7}$ using maximum-entropy analysis

S. Alves, C. Boekema, C. Halim, J. C. Lam, and E. Whang
Physics Department, San Jose State University, San Jose, California 95192-0106

D. W. Cooke and M. Leon
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 26 July 1993; revised manuscript received 19 November 1993)

A maximum-entropy method has been applied to transverse-field muon-spin-rotation vortex data of $R\mathrm{Ba_2Cu_3O_7}$ (R 1:2:3:7; R = Er, Gd, and Eu); this produces spectra representing estimates for the magnetic-field distributions. The information on the field distribution is of better quality than that resulting from Fourier transformation and curve fitting of the same data. Significant deviations from Abrikosov predictions have been observed for the estimated R 1:2:3:7 vortex-field distributions. Below 10 K, for Er 1:2:3:7 and Gd 1:2:3:7, the non-Abrikosov features appear to be influenced by the magnetism of the rare-earth layers.

Muon-spin-rotation (μSR) studies[1] are making significant contributions towards understanding the vortex states of the CuO-based high-T_c superconductors. Improvement in basic understanding of these mixed magnetic and superconducting states is necessary for both fundamental and technological reasons. Whether the field distribution is best described by a vortex glass or a vortex lattice is a crucial issue[2] to be resolved. In the formation of the vortex state, thermal fluctuations and magnetic disorder may play significant roles. Detailed microscopic measurements (such as by μSR) or vortex field distributions can provide direct answers to these fundamental questions.

In most μSR studies of field distributions,[1] the observed muon-decay time histograms are analyzed by Fourier-transformation methods and curve fitting. In principle, the field distribution is given by the *real part* of the Fourier Transform (FT).[3] For real μSR data, which is discrete, noisy and truncated after a few muon-life times, FT analysis is problematic. In several FT and curve-fitting investigations, Gaussian distributions have been reported rather than the discontinuous distributions with sharp features predicted by canonical Abrikosov theory.

An alternative approach for analyzing time series is the maximum-entropy (ME) method.[4] Some particular ME methods, like the Burg algorithm, are based upon autoregression prediction techniques.[4(b)] In contrast to FT, ME is quite successful for short-time range and noisy data, and does not suffer from truncation effects, like the well-known FT sinc wiggles. ME has the major advantage of producing in the frequency spectrum *only structure for which sufficient statistical evidence is present* in the time series. In recent years, ME has been successfully applied to NMR spectroscopy.[4] We have developed and successfully tested an ME method for transverse field (TF) μSR data based on the Burg algorithm.[4(b)]

In this paper, we report the results of our ME analysis of μSR vortex data reported earlier[5] for $R\mathrm{Ba_2Cu_3O_7}$ (R 1:2:3:7; R = Eu, Gd, Er). Below, the relevant μSR vortex studies and theoretical predictions are discussed briefly, followed by an outline of our ME-μSR method. Next, the results of the ME-Burg analysis are presented and discussed. Finally, the conclusions are offered; these results help improve the understanding of the vortex states of cuprate superconductors.

Following the Bednorz and Mueller discovery (1986), several TFμSR studies on CuO-based superconductors have been reported.[1] In standard time-differential μSR techniques, polarized positive muons are implanted one at a time into the sample under study. The time intervals between μ^+ implantation and the decay positron emission (preferentially along the direction of the muon spin) are recorded. The resulting time histogram of μ-decay events is given by $N(t)=N_0\exp(-t/\tau_\mu)[1+A_s(t)]$, where τ_μ is the muon-life time ($\sim$2.2 μsec). When a single magnetic field value is dominant, the time-dependent asymmetry, $A_s(t)$, can be approximated by $A_s(t)\sim A\cdot G(t)\mathrm{cost}(\omega t+\phi)$, where A is the initial asymmetry, $A\cdot G(t)$ is the envelope of the oscillations, ω is the mean precessing angular muon frequency ($\omega=2\pi\nu=2\pi gB$ with $g=13.55$ MHz$\cdot$kOe^{-1}), and ϕ is a phase angle. $A_s(t)$ contains the *exact* information on the magnetic field distribution, while $G(t)$ provides an approximate measure. This information on the probability distribution of the local magnetic fields at the μ^+ stopping sites can be extracted from $A_s(t)$ by means of Fourier transformation, or by maximum-entropy analysis—as reported here in some detail.[6]

In high-T_c cuprates, TFμSR has been successful in the determination of magnetic penetration depths and the strengths of the flux pinning.[1,5,7] Further, it has been claimed that for the 1/2/3 cuprates and other type-II superconductors, T_c is inversely proportional to the square of the $T\rightarrow 0$ London penetration depth.[7] Truncation effects and statistical noise make FT and curve fitting rather insensitive to sharp features of the field distribution. Another (albeit minor) problem is the fact that the muon does not randomly sample the magnetic environment at the atomic scale, as has been frequently assumed. Below about 150 K the muon primarily probes the vicinity of the oxygens in the BaO layers,[8] an *insulating* part of R 1:2:3:7. These difficulties limit studying details of the flux configurations and dynamics.

Several TFμSR line shape studies of type-II superconductors in the mixed state have been published,[1(e),9] following the pioneering work of Ivanter and Smilga.[3(a)] The predicted line shape of the field distribution for a sintered powdered sample exhibits a sharp peak occurring slightly below the applied field value, with a long tail (up to 150 Oe) at higher fields. Over a 50-Oe field interval below the sharp peak, the field distribution initially falls precipitously followed by a steady decline to zero. (See also inset of Fig. 2.) For Y 1:2:3:7, differences between experimental FT shapes and theoretical curves were discussed and explained in terms of flux-trapping models.[1(e)]

Particular aspects of the data acquisition and analysis need to be pointed out. First, in the μSR vortex data[1(b),1(d),(5)] taken at LAMPF, *two* μSR signals are observed: one that broadens drastically with decreasing temperature below T_c, and a second frequency *nearly* equal to that corresponding to the applied field. The LAMPF TFμSR data need *no* "background" subtraction, because the positron counters are placed inside the cryostat, precluding any background signal arising from nonsample sources. This has been confirmed by auxiliary TFμSR measurements on magnetite.[1(d)] This slightly temperature-dependent signal, observed near the applied field, may stem from grain boundaries or other nonsuperconducting regions, where disorder in the chain layers exists at the atomic scale.[1(d)] Second, for the R 1:2:3:7 data, the $A_s(t)$ behavior indicates that A is about 0.21 and below about 60 K, $G(t)$ dies out quickly within 1 μsec. Below T_c, $A \cdot G(t)$ plots show slight, but systematic, deviations from Gaussian behavior near 0 and 1 μsec. Thus, Gaussian curve fitting may overlook significant features of the vortex data.

An ME-μSR method has been developed;[10] a brief description follows. Let x_i be the signal to be analyzed; for μSR this is $A_s(t)$, which is obtained by removing the background noise and muon-decay exponential, and subtracting the nonoscillating term; x_i is sampled in N equally spaced time bins ($i=1,\ldots,N$). One can write[4,11]

$$x_i = \sum_{k=1}^{p} c_k x_{i-k} + n_i \ .$$

The autoregressive coefficients c_k are determined from the data, p is called the order of the model description, and n_i is the assumed white noise term for each bin. Using the Burg minimization criterion to obtain the least structure in the transform, the c_k values are estimated. The power spectral density (PSD) is easily calculated[4] using c_k, and a reliable estimate for the probability distribution is obtained by taking the square root of the PSD;[4(a)] we refer to this as the *ME transform*. For simulated and real TFμSR data in the 10-MHz range, we have determined p empirically by monitoring the Akaike prediction error[11] in this ME approach. The optimal order p_{opt} is about 1/5 to 1/3 of the number of points (N_t) in the time series. Spurious signals appear when p is larger than $\frac{1}{2}N_t$.

To maximize the ME signal-to-noise ratio, the data x_i can be weighted before ME transformation by an exponential or Gaussian filter function with filter time T_f. Taking into account the Poisson statistics of the μ decay, exponential weighting (with twice the μ^+-decay time as T_f) would remove most of the inherent μSR scatter. If the $A_s(t)$ signal shows approximate Gaussian relaxation, a Gaussian filter is more beneficial for signal-to-noise improvement in the ME transform. A $1/T_f$-broadening effect in the ME distributions should be expected. We have found that filtering reduces the background scatter much more at frequencies outside the signal region(s) than where the signals occur.

We have applied our ME method to *simulated* μSR data containing *closely spaced* but discrete frequencies, or having continuous square or triangular frequency distributions, as well as to *actual* ZFμSR data[12] obtained for the antiferromagnetic phase of Y_2BaCuO_5 (Y 2:1:1). All μSR signals are in the 10-MHz range. The simulations show that the ME estimates for the field distributions closely resemble the generated field distributions.[10] ME application to LAMPF ($2M$ statistics) data of Y 2:1:1 confirmed the five signals previously reported and obtained by curve fitting of high statistics PSI data.[13] The precision of all positions of the five *sharp* ME peaks is a factor of 10 better than that of the frequency signals determined by their[13] curve-fitting analysis. During ME development and p_{opt} determination, we saw the need for an estimate of error. We have found two sources of error in the ME transforms: a random noise contribution determined by statistics (as usual), and a smaller one whose value correlates positively with the magnitude of the slope in the ME transform.

We now present and discuss the results of the application of the ME-Burg method to TFμSR data recorded for R 1:2:3:7 in their mixed states. The polycrystalline samples are high-quality, single-phase ceramics R 1:2:3:7 (R=Er, Gd, and Eu) with T_c's of about 94 ($\pm$2) K. These samples were field cooled in a 1-kOe field. (For further details, see our earlier reports.[5]) We note that the Er and Gd ions carry magnetic moments, while Eu (like Y) does not. Er 1:2:3:7 has been reported to be a two-dimensional (2D) Ising antiferromagnet with a T_N of 0.7 K.[14] Gd 1:2:3:7 is known to be a 3D antiferromagnet below T_N of 2.3 K.[15]

In Fig. 1 we show the ME determination of the field distribution in the Er 1:2:3:7 vortex state at 4.5 K. The $A_s(t)$ data used for this transform has been weighted by a Gaussian filter with T_f=0.7 μsec; the signal-to-noise ratio is a maximum for this filter time. Most of the field distribution is well below the applied field, where a small peak can be seen. This contribution may come from nonsuperconducting regions (see above). A "fall-off" occurs about 250 Oe below the main peak, and is followed by a long tail down to 100 Oe. The steep decrease is reminiscent of Abrikosov features for single crystal samples, but is *not* expected in polycrystalline ceramics.[1(e),9] As discussed below, the fall-off in the field distribution appears to be due to the magnetism generated by the Er planes.

These three non-Abrikosov features (the small contribution near the applied field, the fall-off and tail at lower fields) do *not* show up for ME estimates for $Pb_{0.9}In_{0.1}$, a

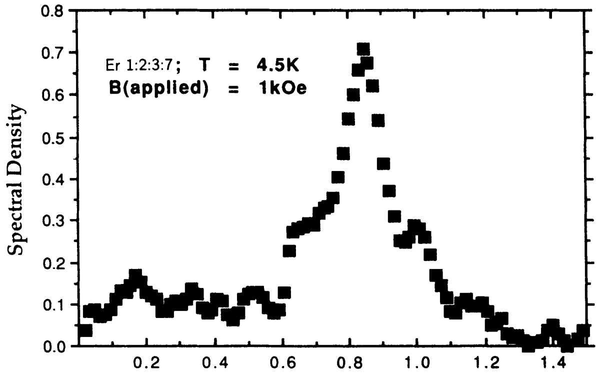

FIG. 1. ME estimate of the field distribution for Er 1:2:3:7 at 4.5 K. The Gaussian filter time T_f used to weight the A_s date is 0.7 μsec, which is about the lifetime of the vortex μSR signal. The error in spectral density is about 0.05, a preliminary conservative estimate (see text). Near the applied field (1 kOe), a contribution from "normal" regions can be seen [Ref. 1(d)]. The main part of the field distribution is at 800 Oe; a "fall-off" is observed near 620 Oe, together with a long tail reaching even lower fields.

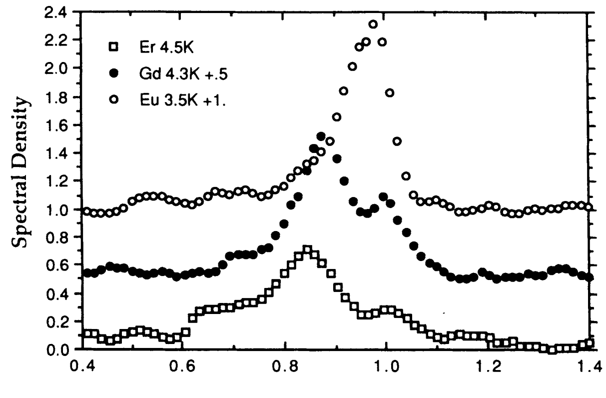

FIG. 3. ME estimates for the field distributions for R 1:2:3:7 (R=Eu, Gd, and Er) at liquid-He temperatures (T_f =0.7 μsec). The applied field is 1 kOe. Vertical off-sets for Gd (+0.5) and Eu (+1.0) have been introduced for clarity. The field distributions appear to depend on the magnetism present in the R planes. Significant deviations from the Abrikosov prediction can be noticed.

typical type-II superconductor, at 3 K and, 500 and 800 Oe.[16] Outside the main field distribution (with a width of 175 Oe, T_f=1.5 μsec), *no* contributions at lower field are seen within error. This indicates the R 1:2:3:7 cuprates are *not* typical type-II superconductors.

In Fig. 2, we compare the ME field distribution estimates for Eu 1:2:3:7 at three different temperatures; well below, below and above T_c. At 150 K, the FWHM of the peak is 40 Oe, which corresponds to 0.54 μsec^{-1}. This can be compared with the estimated broadening effect of the filtering ($1/T_f$=1.4 μsec^{-1}; an exponential filter with T_f=4.4 μsec yields a linewidth slightly smaller than the fitted Gaussian relaxation rate of 0.2 μsec^{-1}). Below T_c (at 80 K), the peak broadens rapidly by a factor of 2, and at 4.6 K the spread is over 200 Oe. No hint of a signal is evident near the applied field, because the $1/T_f$-broadening causes substantial overlap of this small signal with the main vortex signal. Well below T_c, nearly triangular, asymmetric, *non*-Gaussian distributions are observed; deviations from the Abrikosov prediction (see inset Fig. 2) can be noted.

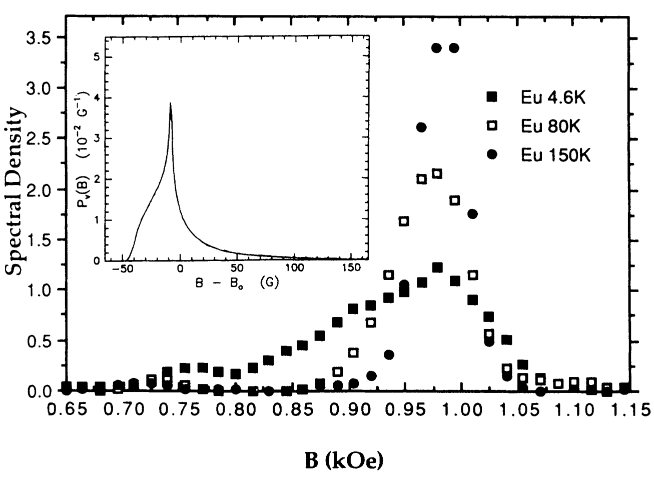

FIG. 2. ME estimates for the field distributions for Eu 1:2:3:7 at 4.6, 80, and 150 K. The applied field is 1 kOe. The 40-Oe width at 150 K is mainly a broadening effect due to Gaussian weighting (T_f=0.7 μsec). The field distribution at 4.6 K is nearly triangular and asymmetric. The inset depicts the Abrikosov prediction for polycrystalline ceramics R' 1:2:3:7 (R' is a nonmagnetic rare-earth ion like Eu or Y: λ_{ab} =1400 Å;λ_c/λ_{ab}=5;κ=100; from Ref. 1(e) and D. R. Harshman (private communication).

In Fig. 3, the ME field distributions for R 1:2:3:7 with R=Er, Gd, and Eu at liquid-He temperatures are compared (T_f=0.7 μsec). A significant fall-off seems *not* to be presented in the distributions for the Eu 1:2:3:7 data recorded at 3.5 K (Fig. 3) and 4.6 K (Fig. 2). The fall-off *is* seen for R=Gd (at 680 Oe) and Er (at 620 Oe), suggesting that perhaps this feature is a magnetic effect. The magnetic field value for the fall-off scales well with the magnetic moment of the R ion. The low-field tail for Eu 1:2:3:7 at 3.5 K running from 0.5 to 0.75 kOe is confirmed by comparison to higher temperatures, where this tail disappears. At 4.6 K a small part of the flat tail centered near 0.77 kOe is still visible (see Fig. 2).

The existence of the low-field tail for all three R 1:2:3:7 $T\rightarrow 0$ field distributions shows that, in the mixed state, regions exist for which the magnetic field is much lower than predicted by Abrikosov theory. Recall[8] that the muons are probing just below the insulating BaO layers. The electrons of the neighboring CuO_2 planes (which are responsible for superconductivity) have managed to screen substantially the applied field for these low-field tail muons. These muons are sensing competing, perhaps random, magnetic interactions. This magnetic frustration appears to be influenced by the presence of magnetic R ions, as can be seen from Figs. 1 and 3. Thus, we speculate that the low-field tail is caused by field-induced magnetic frustration in the vortex-glass[2] state. Further study is clearly needed.

In conclusion, an application of the maximum-entropy technique to TFμSR has been developed and applied to R 1:2:3:7 (R = Er, Gd, and Eu) data reported earlier.[5] These vortex data were recorded in an applied transverse field of 1 kOe at several temperatures below room temperature. The ME information on the field distribution is of better quality than that which resulted from FT and curve-fitting methods on the same data.

Concerning the estimated field distributions in the R 1:2:3:7 vortex states, we have observed significant deviations from Abrikosov predictions. These deviations were *not* seen for the canonical type-II superconductor $Pb_{0.9}In_{0.1}$. For R = Er and Gd, the non-Abrikosov features appear to be influenced by the magnetism of the rare-earth layers. The low-field tail in the vortex field distribution suggests glassy characteristics in the vortex state of cuprate superconductors. ME analysis of other high-T_c cuprate systems measured earlier at LAMPF is underway.

This work was supported by Research Corporation, Associated Western Universities, San Jose State University Foundation, and in part by the U.S. Department of Energy. C.B. thanks the Los Alamos National Laboratory for its kind hospitality. S.A., C.H., J.L.L., and E.W. thank the Society of Archimedes for travel and research support.

---

[1](a) See *Proceedings of the Fifth International Conference on Muon-Spin Rotation,* Oxford, 1990 [Hyperfine Interac. **63-65** (1990)], and references therein; (b) R. L. Lichti *et al.*, Appl. Phys. Lett. **54**, 2361 (1989); (c) B. Pumpin *et al.*, Phys. Rev. B **42**, 8019 (1990); (d) R. L. Lichti *et al.*, *ibid.* **43**, 1154 (1991); (e) D. R. Harshmann *et al.*, Phys. Rev. Lett. **66**, 3313 (1991); (f) D. R. Harshmann *et al.*, Phys. Rev. B **47**, 2905 (1993).

[2](a) D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); (b) M. P. A. Fisher, *ibid* **62**, 1415 (1989); (c) D. R. Nelson and V. M. Vinokur, *ibid.* **68**, 2398 (1992).

[3](a) I. G. Ivanter and V. P. Smilga, Zh. Eksp. Teor. Fix. **55**, 548 (1968) [Sov. Phys. JETP **28**, 286 (1969)]; (b) E. H. Brandt and A. Seeger, Adv. Phys. **35**, 189 (1986).

[4](a) J. A. Jones and P. J. Hore, J. Magn. Res. **92**, 276 (1991); (b) D. S. Stephenson, Progress in NMR Spectroscopy **20**, 515 (1988).

[5](a) D. W. Cooke *et al.*, Phys. Rev. B **37**, 9401 (1988); (b) D. W. Cooke *et al.*, *ibid* **39**, 2748 (1989).

[6]S. Alves *et al.*, APS Bull. **38**, 380 (1993).

[7]Y. J. Uemura *et al.*, Phys. Rev. Lett. **66**, 2665 (1991); **62**, 2317 (1989).

[8](a) C. H. Halim *et al.*, Physica B **163**, 453 (1990), and references therein; (b) C. Boekema, in *Time Domain in Dynamics,* edited by G. Long and F. Grandjean (Kluwer Academic, Dordrecht, 1988), p. 377; (c) W. K. Dawson, Master thesis, San Jose State University, 1991; (d) C. Boekema *et al.* (unpublished).

[9](a) M. Celio *et al.*, Physica C **153-155**, 753 (1988); (b) A. D. Sidorenko *et al.*, Hyperfine Interact. **63**, 49 (1990).

[10]Further details will be reported elsewhere.

[11]T. J. Ulrych and T. N. Bishop, Rev. Geophys. Space Phys. **13**, 183 (1975).

[12]C. Boekema, D. W. Looke, and R. L. Lichti (unpublished).

[13]A. Weidinger *et al.*, Physica C **153-155**, 168 (1988).

[14](a) J. W. Lynn, APS Bull. **38**, 71 (1993); (b) T. W. Clinton *et al.* (unpublished).

[15]H. A. Mook *et al.*, Phys. Rev. B **38**, 12 008 (1988).

[16]Data for $Pb_{.9}In_{.1}$: $T_c = 7$ K; $H_{c1} = 200$ Oe and $H_{c2} = 1.5$ kOe; R. N. Heffner and D.E. MacLaughlin (unpublished).