Search for Anomalous Internal Magnetic Fields in High-T_c Superconductors as Evidence for Broken Time-Reversal Symmetry

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(Received 30 November 1989)

Several recent models of high-T_c superconductivity predict static internal magnetic fields along the c axis due to spontaneously broken parity and time-reversal symmetries. Muon-spin relaxation has been used to study the internal fields in c-axis-oriented samples of sintered $YBa_2Cu_3O_7$ and of thick-film $Bi_2Sr_2CaCu_2O_8$. The magnitude, anisotropy, and temperature dependence of the observed fields favor a nuclear dipolar origin. Any anomalous fields at μ^+ sites in bulk CuO_2 superconductors must be $\lesssim 0.08$ mT.

PACS numbers: 76.75.+i, 74.70.Vy, 75.50.Ee

In spite of intense theoretical and experimental efforts to understand high-T_c superconductivity, many fundamental questions remain unanswered. In particular, it is still not known whether the symmetry properties of the superconducting ground state are those of a conventional superconductor. Laughlin and co-workers[1,2] have proposed a new mechanism for superconductivity in two dimensions involving a gas of quasiparticles called *anyons* which obey fractional statistics. Layered CuO_2 materials, in which the superconductivity is highly two dimensional in nature, are a potential realization of such a model. A distinctive feature of the anyon models is that both time-reversal ($\mathcal{T}$) and parity ($\mathcal{P}$) symmetries are broken at or above T_c while the product $\mathcal{PT}$ is conserved.[3,4]

The experimental consequences of broken $\mathcal{T}$ symmetry in the context of high-T_c superconductors have recently been pointed out by Halperin, March-Russell, and Wilczek.[5] In particular, since the anyons carry a real magnetic moment, they should produce small internal magnetic fields oriented perpendicular to an isolated CuO_2 plane with an estimated magnitude of ~ 3 mT.[6] If the coupling between planes were ferromagnetic, such that $\mathcal{P}$ and $\mathcal{T}$ had the same sign in adjacent planes, there would be observable bulk effects. If, on the other hand, adjacent CuO_2 planes have opposite signs of $\mathcal{P}$ and $\mathcal{T}$ (Ref. 7), the effects should only be seen on surfaces or microscopically within layers. For example, internal magnetic fields with a reduced magnitude should be found at most crystalline sites due to the periodic variation of the anyon density within a CuO_2 plane.[6] In fact, the diamagnetic currents responsible for the anyon magnetic moment are a feature of many mean-field theories of the Hubbard model. They were first predicted to occur in "flux phases."[8]

Muon-spin relaxation[9] (μSR) is a powerful method for detecting the resulting internal magnetic fields. Screening of the positive charge of the muon should further perturb the local anyon density from its average value, assuring some finite internal magnetic field[6] provided the muon is not located precisely at sites where such fields cancel by symmetry. The same argument also applies to flux-phase theories. In this Letter we report precise μSR measurements of the internal magnetic fields in c-axis-oriented $YBa_2Cu_3O_7$ sintered powder and $Bi_2Sr_2CaCu_2O_8$ thick film and in an unoriented finely ground powder. We conclude that the observed fields are not ascribable to anyons and that the magnitude of any anomalous magnetic fields along the c axis in bulk material must be $\lesssim 0.08$ mT.

The oriented sample of sintered $YBa_2Cu_3O_7$ ($T_c = 90$ K) was prepared at the General Electric Corporation. A neutron-scattering rocking curve indicated that the c axes of the powder grains were aligned to within about ±9°. Prior to the experiment the sample was heated to 700 °C in an atmosphere of purified dry oxygen and then slowly cooled to room temperature over a period of two days to ensure complete oxygenation. The $Bi_2Sr_2CaCu_2O_8$ film ($T_c = 85$ K) was prepared at the University of British Columbia (UBC) starting with multiply reacted $Bi_2Sr_2CaCu_2O_8$ powder spread on cleaved MgO. The sample was heated to 1300 °C (well above the melting temperature), cooled slowly to 890 °C at a rate of 60 °C/h, held at that temperature for 16 h, and then furnace cooled to room temperature. X-ray diffraction showed the resulting film to be 95% single phase with the c axes highly oriented perpendicular to the face. Torque measurements confirmed that the c axes were oriented to within ±10°. The fine-grained $YBa_2Cu_3O_7$ sample was prepared at UBC starting from a stoichiometric mixture of 99.999% pure Y_2O_3, $BaCO_3$, and CuO. After the reaction the fully oxygenated single-phase $YBa_2Cu_3O_7$ material was ground with a high-energy vibro ball mill so that the particle diameter ranged from 200 to 1000 nm (about 100 times smaller than for the sintered sample). The powder x-ray-diffraction pattern of the ground material was consistent with fully oxygenated $YBa_2Cu_3O_7$ plus line broadening due to particle-size effects. Prior to the experiment the samples were stored in a dry atmosphere.

In the experiment, conducted on the M20B beam line at TRIUMF, spin-polarized surface muons were collimated to a 12-mm-diam beam spot and then passed through a thin muon detector before entering a He-gas flow cryostat. Over 90% of the muons were stopped in the samples, which covered an area of $\gtrsim 4\ \mathrm{cm}^2$. For the oriented samples the c axes were aligned parallel to the beam direction. A Wien filter, consisting of crossed electric and magnetic fields, was used to rotate the spin polarization of the muon beam either parallel or perpendicular to the c axis without disturbing the sample or other beam characteristics. Stray fields at the sample position were cancelled to within 0.01 mT in all directions.

The spin of the implanted μ^+ acts as a sensitive probe of static internal magnetic fields, capable of detecting fields as small as 0.01 mT.[10-14] As a result of parity violation in muon decay, the muon-spin polarization can be monitored without the use of externally applied magnetic fields.[9] This zero-field (ZF) μSR technique[15] is particularly useful in the present experiment since it can be performed equally well above and below T_c without interference from supercurrents and the resulting magnetic fields generated in response to an external field.

Suppose there exists a small local internal magnetic field directed along the $\pm\hat{\mathbf{c}}$ axis due to anyons and that the initial direction of muon polarization is along $\hat{\mathbf{z}}$. Then for times t much less than the period of muon precession the component of the muon polarization along the initial direction of polarization $\hat{\mathbf{z}}$ evolves in time according to

$$G_{zz}(t) \sim \exp(-\Delta^2 t^2)\,, \tag{1}$$

where

$$\Delta^2/\gamma_\mu^2 = \tfrac{1}{2}(\langle B_x^2\rangle + \langle B_y^2\rangle) \tag{2}$$

is half the mean-squared internal field in the x-y plane perpendicular to $\hat{\mathbf{z}}$ and $\gamma_\mu = 0.852\times10^6\ \mathrm{s^{-1}\,mT^{-1}}$. Note that Δ^2 should vanish when $\hat{\mathbf{z}}$ is parallel to the c axis and be a maximum when $\hat{\mathbf{z}}$ is perpendicular to the c axis.

Previous ZF-μSR measurements[14] below T_c indicate that the rms magnitudes of the internal magnetic fields in $YBa_2Cu_3O_x$ ($x = 6.74$) and $La_{1.85}Sr_{0.15}CuO_4$ are not much larger than about 0.3 mT or about 10% of the estimated "anyon field" for an *isolated* plane. However, this is still consistent with an anyon interpretation since the planes are not isolated and there are uncertainties in both the muon location and the local variation of the anyon density around the muon. We report below a precise measurement of the orientation and temperature dependence of these internal fields. Our results strongly support an interpretation of these fields involving muon-nuclear dipolar interactions over one involving anyons.

The largest abundant nuclear moments in $YBa_2Cu_3O_7$ are those of ^{63}Cu (69.1% abundant, $J = \frac{3}{2}$, $\mu_N = 0.795\times\mu_p$) and ^{65}Cu (30.9% abundant, $J = \frac{3}{2}$, $\mu_N = 0.852\mu_p$). The nuclear moment of ^{39}Y (100% abundant, $J = \frac{1}{2}$) is about 15 times smaller. In $Bi_2Sr_2CaCu_2O_8$, ^{209}Bi (100% abundant, $J = \frac{9}{2}$, $\mu_N = 1.45\mu_p$) has a nuclear moment about 1.8 times that of Cu. In the presence of an electric-field gradient (and resulting nuclear quadrupole interaction[16]) nuclei such as Cu and Bi exert a static classical dipolar field on the muon:

$$\mathbf{B}_{\mathrm{dip}} = \hbar\gamma_n J_q[3(\hat{\mathbf{r}}\cdot\hat{\mathbf{q}})\hat{\mathbf{r}} - \hat{\mathbf{q}}]/r^3\,, \tag{3}$$

where J_q is the component of nuclear spin along the electric-field-gradient direction $\hat{\mathbf{q}}$, and $\hat{\mathbf{r}}$ is the unit vector between the muon and the nucleus separated by a distance r. In a complex crystal structure like $YBa_2Cu_3O_7$ or $Bi_2Sr_2CaCu_2O_8$ where there may be multiple muon stopping sites,[12,14] each with possibly several near-neighbor nuclei, one expects a broad and nearly isotropic distribution of nuclear dipolar fields. Assuming a Gaussian distribution of internal fields with random orientation leads[15] to a muon polarization function

$$G_{zz}(t) = \tfrac{1}{3} + \tfrac{2}{3}(1 - \Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2)\,, \tag{4}$$

which in the low-field limit ($\Delta t \ll 1$) approaches a simple Gaussian form [see Eq. (1)], where Δ is defined in Eq. (2).

Our ZF-μSR time spectra (see, for example, Fig. 1) were fitted by Eq. (4) multiplied by an instrumental asymmetry factor (A) which depends on the properties of muon decay and the positron detectors. The fitted values of $\Delta_\parallel$ (obtained with the muon polarization parallel to the c axis) and $\Delta_\perp$ (obtained with the muon polarization perpendicular to the c axis) in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ are shown in Figs. 2(a) and 2(b), respectively. Using Eq. (2) the rms field components parallel and perpendicular to the c axis can be derived from $\Delta_\parallel$ and $\Delta_\perp$:

$$\langle B_\parallel^2\rangle^{1/2} = (2\Delta_\perp^2 - \Delta_\parallel^2)^{1/2}/\gamma_\mu\,, \tag{5}$$

$$\langle B_\perp^2\rangle^{1/2} = \Delta_\parallel/\gamma_\mu\,, \tag{6}$$

from which one can calculate the rms field magnitude.

The most noteworthy features in the data shown in

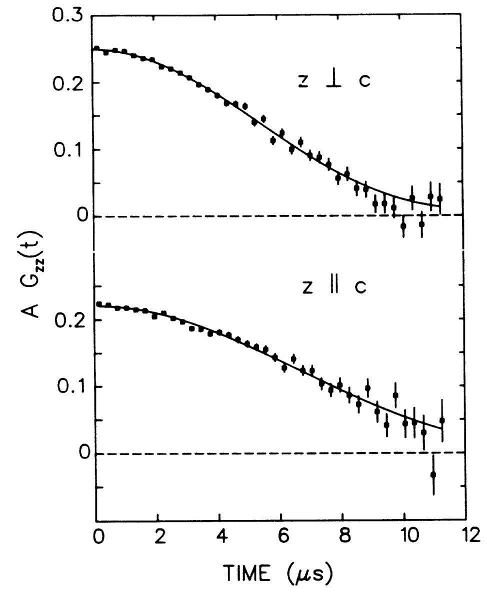

FIG. 1. Time evolution of the muon polarization in zero external field for a c-axis-oriented sintered powder of $YBa_2Cu_3O_7$ at $T=6$ K with the initial muon polarization ($\hat{z}$) parallel and perpendicular to the c axis.

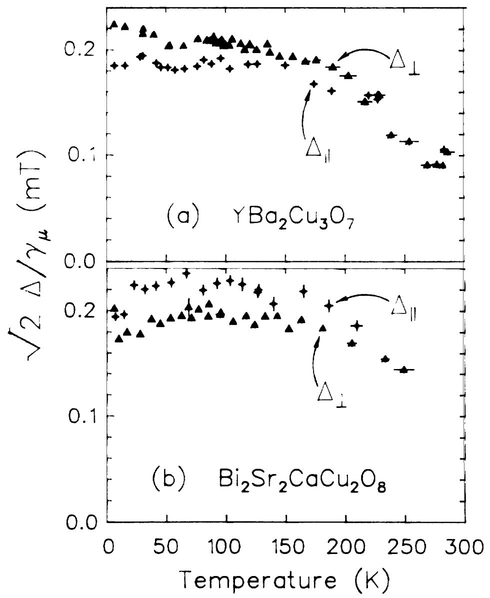

FIG. 2. (a) The measured linewidths $\Delta_\parallel$ and $\Delta_\perp$ in c-axis-oriented $YBa_2Cu_3O_7$ sintered powder for the initial muon polarization parallel and perpendicular to the c axis, respectively. These are related to the rms components of the internal field by Eqs. (5) and (6). (b) The same for thick-film $Bi_2Sr_2CaCu_2O_8$.

Figs. 2(a) and 2(b) are the weak orientation dependence of the internal fields, the near equality of the magnitude of the fields in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$, and the gradual decrease of their apparent magnitude with increasing temperature. From Fig. 2(a) and Eqs. (5) and (6) one finds in $YBa_2Cu_3O_7$ that the rms magnitude of B is 0.25 mT and that $\langle B_\parallel^2\rangle^{1/2}$ is 15(3)% larger than $\langle B_\perp^2\rangle^{1/2}$. If there were no anisotropy in the nuclear dipolar fields then one could conclude that there was an anomalous field in $YBa_2Cu_3O_7$ along the c axis with an rms value equal to about 0.08(2) mT. (This assumes that the rms components from nuclear dipoles and other origins add in quadrature.) However, the opposite anisotropy is seen in $Bi_2Sr_2CaCu_2O_8$ [see Fig. 2(b)] where $\langle B_\parallel^2\rangle^{1/2} < \langle B_\perp^2\rangle^{1/2}$; thus the observed small anisotropies are probably due to the anisotropic nature of the muon-nuclear dipolar interaction.

The near equality of the internal fields in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ is noteworthy considering the different and complex crystal structures and the fact that the dipolar fields vary as the cube of the muon-nuclear distance [see Eq. (3)]. One explanation is that in both cases the muons are close to the common CuO_2 planes so that the μ^+-Cu nuclear dipolar interaction is dominant and about equal in the two materials. This is supported by recent μSR and muon level-crossing experiments[17] on $YBa_2Cu_3O_7$ containing 38% ^{17}O ($S=\frac{5}{2}$, $\mu_N=0.678\mu_p$). These results indicate that the muon is located 1.0 Å away from an oxygen ion and that the electric-field-gradient tensor on that oxygen is very close to what is measured by NMR for oxygen in the CuO_2 plane[18] and well away from that seen for the chain-site oxygens. This confirms that the muon forms a μ^+-O bond as calculations indicate[19] but does not support the site assignment in $YBa_2Cu_3O_7$ proposed by Birrer *et al.*[20] in which the muon is located 1 Å away from an oxygen in the CuO chain. Note that from symmetry considerations one would expect the internal fields due to anyons to be largest for muon sites near a CuO_2 plane and cancel to zero for sites midway between two such planes with opposite signs of $\mathcal{P}$ and $\mathcal{T}$. It should be noted that no studies so far indicate that the muon is exactly between two CuO_2 planes (e.g., in the plane of the CuO chains), and that our upper limit on the anyon contribution to the internal field is about 30 times smaller than that estimated for a muon in an isolated CuO_2 plane.

The temperature dependence of the internal fields is also consistent with nuclear dipolar fields, although there are some small anomalies. Note that there is no detectable change in the internal fields at T_c, implying that the magnitude of any anomalous magnetic field appearing at T_c is $\lesssim 0.03$ mT. The gradual decrease in $\Delta_\parallel$ and $\Delta_\perp$ above 200 K is consistent with motional averaging of the muon-nuclear dipolar interaction as the muon begins to hop between sites. The small variations in $\Delta_\parallel$ and $\Delta_\perp$ in $YBa_2Cu_3O_7$ below 100 K are not yet understood but are unlikely to be connected with anyons since they are similar for both orientations of the muon polarization and since the same effect is not seen in $Bi_2Sr_2CaCu_2O_8$.

We also studied a sample of unoriented $YBa_2Cu_3O_7$

with a very fine grain size of about 200-1000 nm in order to see if the internal fields within a few London penetration depths[21] (~140 nm) of the surface differed appreciably from those in the bulk below T_c. Although the fitted linewidth parameter Δ was close to that in the sintered $YBa_2Cu_3O_7$ above 150 K, the fits by Eq. (4) were relatively poor and became increasingly worse at lower temperatures. At 6 K an excellent fit to the data was obtained with $G_{zz}(t)=\exp(-\lambda t)$ with $\lambda=0.01$ μs^{-1}, indicating a Lorentzian distribution of internal fields. The high-field components necessary to produce such a $G_{zz}(t)$ cannot be explained by nuclear dipole moments. The origin of these fields may be due to a dilute concentration of Cu electronic moments resulting from surface contamination or damage incurred during the grinding process.

In conclusion, μSR measurements of the internal magnetic fields in oriented samples of $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ indicate that the rms magnitude of the internal magnetic fields in bulk material is about 0.25 mT. The weak orientation and temperature dependence of the internal fields are consistent with dipolar interactions between the muon and Cu and/or Bi nuclear moments. If there are any internal magnetic fields specifically due to breaking of time-reversal invariance at μ^+ sites in bulk material, they are less than about 0.08 mT.

This work was supported by the Canadian Institute for Advanced Research, the National Research Council, the Natural Sciences and Engineering Research Council of Canada, the U.S. National Science Foundation under Contract No. DMR-89-13784, and the David and Lucile Packard Foundation. We would like to thank Keith Hoyle and Curtis Ballard for technical support.

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