## Evidence for Static Magnetism in the Vortex Cores of Ortho-II YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub>

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Evidence for static alternating magnetic fields in the vortex cores of underdoped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> is reported. Muon spin rotation measurements of the internal magnetic field distribution of the vortex state of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> in applied fields of H = 1 T and H = 4 T reveal a feature in the high-field tail of the field distribution which is not present in optimally doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.95</sub> and which fits well to a model with static magnetic fields in the vortex cores. The magnitude of the fields is estimated to be 18(2) G and decreases above T = 10 K. We discuss possible origins of the additional vortex core magnetism within the context of existing theories.

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The application of a large magnetic field to a superconductor drives part of the sample, the vortex cores, into a "normal" state. While the physics of these vortex cores in conventional superconductors is generally thought of as metallic, the vortex cores of high temperature superconductors may offer insight into more unusual low temperature properties. A new class of theories has predicted that magnetism may be induced near or inside the cores of vortices by the application of a magnetic field [1-7]. For example, in Zhang's SO(5) theory, static antiferromagnetism (AF) should appear in regions where the superconducting (SC) order parameter is suppressed, and, as a consequence, vortices in underdoped superconductors should be magnetic [1,2]. Similarly, Wen and Lee have predicted a staggered flux phase (SFP) of orbital currents in the vortex cores of underdoped cuprates [3,4]. Below  $T_c$ , the system is locally a *d*-wave superconductor away from the vortex cores, but has a SFP "frozen" inside the vortex cores. Consequently, they predict the appearance of quasistatic alternating magnetic fields of order 10 G in the vortex cores. Finally, Zhu and Ting have developed the Hubbard model with an on-site repulsion term and shown that AF-like spin density waves (SDW) can appear in the vortex cores [5].

The possible coexistence of magnetism and superconductivity in the cuprates has been a key issue since the discovery of high- $T_c$  superconductors. In zero field, microscopic coexistence of superconductivity, magnetism, and spin-glass behavior has been detected with  $\mu$ SR in the underdoped region near the boundary between SC and AF [8], in Ca-doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> [9], La<sub>2- $\delta$ </sub>Sr<sub> $\delta$ </sub>CuO<sub>4</sub> [9,10], and underdoped and optimally doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> [11,12]. There is also recent experimental evidence from neutron scattering for anomalous magnetism in underdoped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> [13,14] in zero applied field. Field-induced low-frequency magnetic fluctuations in La<sub>2- $\delta$ </sub>Sr<sub> $\delta$ </sub>CuO<sub>4</sub> have been reported by Lake *et al.* [15] and in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> by Mitrovic *et al.* [16]. Vaknin *et al.* [17] found a possible signature of AF cores in optimally doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub>, while Katano report sharp incommensurate neutron scattering peaks in  $La_{2-\delta}Sr_{\delta}CuO_4$  that are enhanced in a magnetic field [18].

In this paper, we report a search for static magnetism in the region of the vortex cores of underdoped Ortho-II YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub>. Significant improvements of the fits to the muon spin precession signal in the vortex state are obtained using a model of the vortex lattice with an additional alternating magnetic field of 18(2) G, whose magnitude decreases away from a vortex core center on the length scale of the coherence length. As a control, similar fits were made on data from the conventional superconductor NbSe<sub>2</sub> in which case no improvement in the fits was observed, as expected.

Muons are an excellent probe of magnetism in superconductors. As described elsewhere [19], the implanted spin polarized muons stop randomly on the length scale of the vortex lattice and precess at a rate proportional to the local magnetic field, thus providing a direct measure of the local field distribution n(B). The observed asymmetric magnetic field distribution in a superconductor is characteristic of a lattice of magnetic vortices in a type-II superconductor. In Ginzburg-Landau (GL) theory, a vortex core's size is determined by the applied magnetic field H and the in-plane GL coherence length  $\xi_{ab}$ , while the magnetic field decays away from the vortex core over a length scale given by the GL ab in-plane penetration depth  $\lambda_{ab}$ . The magnetic field distribution n(B) can be calculated from the spatial distribution of the magnetic field [20]:

$$B(r) = \frac{\Phi_0}{S} (1 - b^4) \sum_G e^{-i\vec{G}\cdot\vec{r}} \frac{uK_1(u)}{1 + \lambda_{ab}^2 G^2}, \quad (1)$$

where  $u^2 = 2\xi_{ab}^2 G^2 (1 + b^4) [1 - 2b(1 - b)^2]$ ,  $K_1(u)$  is a modified Bessel function,  $\vec{G}$  is a reciprocal lattice vector of the vortex lattice,  $b = H/H_{c_2}$  is the reduced field,  $\Phi_0$  is the flux quantum, and S is the area of the reduced unit cell for a hexagonal lattice.

A phenomenological model of n(B) that includes alternating fields in the vortex cores can be created by adding a term to Eq. (1):

$$B'(r) = B(r) + (-1)^{(x+y)/a} M e^{-(r/\xi_{ab})^2},$$
 (2)

where M is the amplitude of the alternating fields in the vortex core that alternates in sign between neighboring crystal lattice sites (a is the distance between sites). Our model assumes that the amplitude of the fields in the vortex cores decays away from the vortex core center over the same length scale  $\xi_{ab}$  as the SC order parameter increases. We note the following: (1) Equation (2) strictly holds only in the unit cell of the vortex lattice, in the limit  $\xi_{ab}/L_0 \ll 1$ , where  $L_0$  is the distance between vortex cores. This ensures there is negligible overlap between alternating fields of neighboring vortices. (2) Because the vortex and crystal lattices are incommensurate, the parameter a has been varied between the Cu-Cu distance in the planes and a much smaller value that allows the muon to experience all alternating fields from -M to M. The results presented here are insensitive to such a change.

Figure 1 shows the effect of the vortex core alternating fields on n(B). The dashed line shows n(B) without alternating fields (M = 0 G), for typical values  $\lambda =$ 2450 Å and  $\xi = 35$  Å in an applied field of H = 4 T. The high-field cutoff corresponding to the magnetic field at the vortex core center is visible at b. The modified n(B) due to the introduction of the fields of magnitude M = 15 G (solid line) shows a double step in the highfield tail at a and c. The lower step (at a) is caused by additional fields in the vortex core antiparallel to the applied magnetic field H, whereas the upper step (at c) originates from sites where the additional fields are aligned with the applied field. The spatial variation of the magnetic field near the vortex cores is shown in the inset. The line corresponds to B(r) from Eq. (1) without alternating fields, while the squares show B'(r) from Eq. (2) with the fields. The Van Hove singularity in n(B) at a is caused by the nearly flat field distribution in the inset along the lower line of square points. These points correspond to fields aligned opposite the applied field.

With a model for n(B), a theoretical function of the muon spin precession signal  $P(t) = P_x + iP_y$  is constructed from Eq. (1) or (2) by calculating the local magnetic field at points on a lattice in the reduced unit cell and summing the contributions from all sampling points:

$$P_x(t) = P(0)G(t) \int_0^\infty n(\omega) \cos(\omega t + \delta) d\omega , \quad (3)$$

where  $\omega = \gamma_{\mu} B$ , G(t) is a Gaussian relaxation function that accounts for additional broadening due to nuclear dipole moments and lattice disorder [21], and  $2\pi\gamma_{\mu} =$ 135.54 MHz/T.



FIG. 1. Magnetic field distribution n(B) in superconductors in an applied field of H = 4 T, with and without vortex core alternating fields. Solid line: n(B) calculated from Eq. (2)with alternating fields in the vortex cores (M = 15 G); dashed line: field distribution without alternating fields calculated from Eq. (1) with identical values for  $\xi$  and  $\lambda$ . Notice that significant changes occur only in the high-field tail. See text for further details. Inset: B(r) along a line between two vortex cores. The solid line shows B(r) without alternating fields, whereas the squares show B(r) with M = 15 G alternating fields in the vortex core. The magnitude of the alternating fields is assumed to decay away from the vortex core center on the length scale of  $\xi_{ab}$ .

In our fits to the muon spin precession signal, n(B) parameters  $\xi_{ab}$ ,  $\lambda_{ab}$ , M, and H were allowed to vary. It has been shown previously that Eqs. (1) and (3) can be used to model the muon spin precession signal in NbSe<sub>2</sub> [22,23] and YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> [24,25] superconductors. In this work, constraints on the fits of Eq. (3) were applied to the range of allowed values of  $\xi_{ab}$ . We find that at the highest applied fields (H = 4 T),  $\xi_{ab} \approx 25$  Å, a value smaller than expected from previous  $\mu$ SR measurements of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.60</sub>. Therefore, in the fits,  $\xi_{ab}$  is constrained to  $\xi_{ab} > 35$  Å at H = 4 T. Unconstrained fits yield *larger* values for the alternating field magnitude.

The  $\mu$ SR experiments were performed on the M15 surface muon channel at TRIUMF using a horizontal gas-cooled cryostat and a high-field (7.5 T) high timing resolution spectrometer (BELLE). All data were taken under conditions of field cooling, and high statistics runs were made with approximately  $30 \times 10^6$  muon decay events for each value of *B*, *T*. A mosaic of high purity YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> ( $T_c = 59$  K,  $\Delta T_c = 0.6$  K) single crystals were used. They were grown in BaZrO<sub>3</sub> crucibles, and detwinned below 200°C. Ortho-II ordering was achieved by low temperature annealing for 7 days [26]. The oxygen content of the sample was chosen due to its stoichiometry and high degree of crystalline order, proximity to the AF/SC boundary, and the oxygen ordering of the Ortho-II phase.

 $\mu$ SR measurements performed in zero magnetic field in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> show no internal magnetic fields. Furthermore, the magnetic field dependence of the skewness and

second moment of the field distribution shows no signature of a transition to a 2D pancake state [27] at T = 2.2 K, where n(B) is no longer described by Eqs. (1) and (2). In a field of 4 T, an asymmetric field distribution, the signature of a well-ordered lattice, is visible up to T = 30 K. Further details of the vortex phase diagram in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> will be published elsewhere.

Figure 2 shows a fit to the muon spin precession signal in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> assuming the phenomenological field profile of Eq. (2). Notice the excellent fit of the theoretical line to the data, and the small uncertainty in the data due to the high statistics. The significant improvement in the fits of our model with alternating magnetic fields is shown in Fig. 3. Figure 3(a) shows the deep minimum in  $\chi^2$ /NDF (number of degrees of freedom) achieved by the addition of fields of magnitude M = 15 G. Figures 3(b) and 3(c) show that a similar effect is not found in optimally doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.95</sub> (at H = 6 T) or in the conventional superconductor NbSe<sub>2</sub> (at H = 0.7 T), which is generally well described by BCS theory. The absence of this effect in these materials indicates that the improvement of the fits is not simply due to the addition of another parameter and that the feature in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> is not present in optimally doped YBa2Cu3O6.95 or conventional superconductors. The curves in Fig. 3 are guides to the eye.

The improvements in the fit can be seen more clearly in the frequency domain. Fast Fourier transforms (FFT's) of data from Fig. 2 and the corresponding fits are shown in Fig. 4. The triangles are an FFT of the data, whereas the full (dashed) line is a FFT of the fit with (without) vortex core fields. Figure 4(b) shows the high-field tail of the Fourier transforms in greater detail. The improved fit is clearly visible in the high-field tail, despite the broadening introduced by Fourier transforming finite data. Additional weight is present at fields higher than the high-field cutoff found using Eq. (1). Similar improvements in the fits were found at other temperatures in applied fields of H = 4



and 1 T. The amplitude of the magnetic fields M in the vortex core center decreases gradually with temperature above T = 10 K. Beyond T = 20 K, no clear signature of the fields could be seen.

In our experiment, the muon is sensitive to the *c*-axis component of the local magnetic field at the muon site. Estimating the size and direction of the effective magnetic moment responsible for these alternating fields from these results is not easy. However, Hsu et al. calculated that the geometric factor due to the displacement between the muon site and the center of a CuO plaquette would be of order 1 [28], assuming the muon bonds to the oxygen above the CuO planes. Hence measurement of 18 G static fields at the muon site in the center of the vortex core agrees quantitatively with theoretical predictions [2,4]. Various other forms of the alternating field term in Eq. (2) have also been tried. Replacing the Gaussian function in the alternating field term by an exponential yields a worse fit (although  $\chi^2$  is still a minimum at nonzero M), while fitting with a second length scale for the magnetic field amplitude decay function (instead of  $\xi_{ab}$ ) provides only a marginal improvement in the fit.



FIG. 2. Muon polarization signal  $P_x(t)$  in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> in an applied field of H = 4 T and at T = 5 K displayed in a rotating reference frame. The line is a fit to  $P_x(t)$  using Eqs. (2) and (3).

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FIG. 3. The best  $\chi^2$  per degree of freedom versus the amplitude of the alternating field, M: (a) YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> at H = 4 T and at T = 5 K (solid squares); (b) YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.95</sub> at H = 6 T (solid nablas); and (c) for NbSe<sub>2</sub> at H = 0.7 T (open squares). The solid curves are guides to the eye.



FIG. 4. (a) Fast Fourier transform of  $\mu$ SR data (triangles) and fit (solid line) in Fig. 2 and of the best fit to Eqs. (1) and (3) without alternating fields (dashed line). (b) The high-field region in greater detail. Notice the improved fit in the high-field tail due to the alternating fields, corresponding to the vortex core area.

We note that there may be a number of other possible interpretations of the anomalous n(B) we have measured. For example, little is known about vortices in underdoped high- $T_c$  superconductors. The *d*-wave nature of the vortices or changes in the pairing symmetry may affect n(B). However, to our knowledge no theories currently exist that predict a change in the high-field tail due to such effects. Another possibility is that the vortex lattice is characterized by two domains that coexist in the sample, one a wellordered vortex flux lattice and the other a disordered flux lattice. The high quality of the sample makes the likelihood of such coexistence small [26]. Thus, we are led to the conclusion that the most likely interpretation is the presence of alternating fields in the vortex cores. As noted above, numerous theories predict their existence, and experimental evidence of fluctuating AF fields in the vortex cores of  $La_{2-\delta}Sr_{\delta}Cu_2O_4$  has been recently reported in neutron scattering measurements. Our main result is precisely the signature one would expect in the field distribution of a superconductor with additional alternating magnetic fields in the vortex cores.

In conclusion, we have shown that a phenomenological model of the field distribution with alternating magnetic fields in the vortex cores fits the muon spin precession signal in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.50</sub> in high magnetic fields significantly better than a model without such vortex core fields. We find the amplitude of these fields to be 18(2)Gat the muon site. The origin of the anomalous magnetism reported here may be consistent with Zhang's SO(5) spin magnetism or various t - J Hubbard models of orbital magnetism. Further measurements at intermediate doping between x = 0.5 and 0.95 are in progress. This measurement is of significant importance to a class of theories of high- $T_c$  superconductors that predict spin or orbital magnetism in underdoped cuprates.

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