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Determination of anisotropic H_{c2} up to 60 T in $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystals

M. M. Altarawneh, K. Collar, and C. H. Mielke
MPA-NHMFL, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

N. Ni, S. L. Bud'ko, and P. C. Canfield
Department of Physics and Astronomy, Ames Laboratory US DOE, Iowa State University, Ames, Iowa 50011, USA
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The radio frequency penetration depth was measured in the superconductor $(Ba_{0.55}K_{0.45})Fe_2As_2$ under pulsed magnetic fields extending to 60 T and down to 14 K. Using these data we are able to infer a $H_{c2}(T)$, H-T phase diagram, for applied fields parallel and perpendicular to the crystallographic c axis. The upper critical-field curvature is different for the respective orientations, but they each remains positive down to 14 K. The upper critical-field anisotropy is moderate, ≈3.5 close to T_c, and drops with the decrease in temperature, reaching ≈1.2 at 14 K. These data and analysis indicate that (i) $(Ba_{0.55}K_{0.45})Fe_2As_2$ anisotropy diminishes with temperature and has an unusual temperature dependence and (ii) $H_{c2}(T=0)$ for this compound may easily approach fields of 75 T.

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The discovery of relatively high-temperature superconductivity in several families of FeAs-based compounds[1,2] has raised hope in both basic and applied physics circles. Whereas these materials present intriguing questions about mechanism, lying as they do near a confluence of structural and magnetic phase transitions,[1–4] they also hold out the promise of not only high T_c values but also apparently extremely high H_{c2} values as well. RFeAs$(O_{1-x}F_x)$ compounds have been shown to have $H_{c2}(T)$ curves that can be reasonably extrapolated to low-temperature $H_{c2}(0)$ values in excess of 100 T.[5,6] Although in some cases anisotropic H_{c2} data can be inferred from magnetization measurements on polycrystalline samples,[7] to date, the lack of sizable single crystalline RFeAsO samples has prevented thorough determinations of anisotropic $H_{c2}(T)$ curves over extended field ranges.

Fortunately, methods for growing single crystals of superconducting $(Ba_{1-x}K_x)Fe_2As_2$,[8] and subsequently the Sr and Ca analogs, were readily developed based on the fact that the AFe_2As_2 compounds are true intermetallics and are amenable to conventional metallic solution growth techniques. The availability of large single crystals allows for the determination of anisotropic $H_{c2}(T)$ data. Initial anisotropic magnetoresistivity data collected on $(Ba_{0.55}K_{0.45})Fe_2As_2$ show that there is a very minor suppression of T_c even for 14 T.[8] Extrapolations of $H_{c2}(T)$ plots inferred from these data to $T=0$ K suggest that $H_{c2}(T)$ for this discovered superconductor may easily exceed 70 T, and interpolation of the data indicates that the upper critical-field anisotropy parameter, $\gamma=H_{c2}^{\perp c}/H_{c2}^{\parallel c}$, is approximately 3.5 close to T_c. Higher-field data are clearly required in both cases to improve our understanding and to place the extrapolation of $H_{c2}(T)$ on firmer footing.

In this Rapid Communication we present anisotropic high-frequency susceptibility measurements on single crystals of $(Ba_{0.55}K_{0.45})Fe_2As_2$ grown in the same manner as those used to collect the initial $H\leq 14$ T data.[8] We are able to extend $H_{c2}(T)$ curves to significantly higher magnetic fields and lower temperatures. As a result we find that (i) γ varies in a continuous manner from a relative maximum of ≈3.5 near T_c to a value of 1.16 near 14 K and (ii) the extrapolation of the $H_{c2}^{\parallel c}(T)$ and $H_{c2}^{\perp c}(T)$ curves to lower temperature indicates that values between ≈75 and 80 T are likely in this material.

Single crystals of $(Ba_{0.55}K_{0.45})Fe_2As_2$ were grown in precisely the manner described in Ref. 8. For these crystals the potassium content was directly measured by wavelength dispersive x-ray spectroscopy and was found to have an average value of 0.45 with a 0.07 standard deviation from layer to layer.[8]

Radio frequency (rf) contactless penetration depth measurements were performed on the single-crystal sample in a 60 T pulsed field magnet with a 10 ms rise time and a 40 ms extended decay. The rf technique was used as it has proven to be a sensitive (typically ~1 part per 1000 resolution) and accurate method for determining the upper critical field in anisotropic superconductors.[9] A radio frequency probe based on a commercially available integrated circuit (IC) proximity detector was used to establish a high stability tank circuit oscillator. The IC delivers a voltage proportional to the tank circuit voltage. The fundamental resonant frequency is approximately 28 MHz at T_c. The technique is highly sensitive to small changes (~1–5 nm) in the rf penetration depth[9] when the sample is in the superconducting state. As the magnetic field increases, the probe detects the transition to the normal state by tracking the shift in resonant frequency (proportional to the penetration depth, i.e., $\Delta\lambda=\Delta f R^2/f_0 r_s$, where R is the detection coil radii, r_s is the sample radii, f_0 is the fundamental frequency, and Δf is the frequency shift[9]). The IC voltage is mixed with an intermediate frequency (IF), and the difference frequency is extracted by using a low-pass filter at 1.9 MHz. This simple rf technique allows for the frequency shift to be recorded with a digitizing oscilloscope at a rate of 80 ns/point (12.5 MHz) for a duration of 100 ms (1.25 Msamples/channel). A simple peak-finding algorithm runs on the raw data to calculate the frequency shift as a function of time. The time-dependent frequency is then correlated with the applied magnetic field yielding a frequency vs magnetic-field plot (see Figs. 1 and 2).

A small ($0.55\times 0.55\times 0.014$ mm^3) single crystal was chosen as larger (>1 mm) cross-section samples tend to ex-

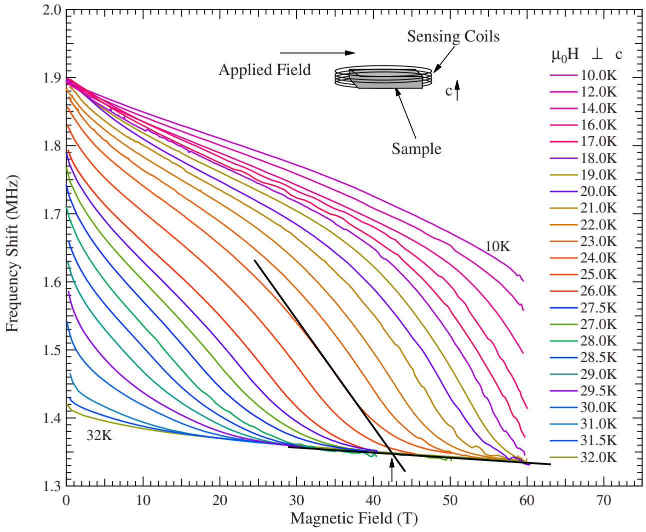

FIG. 1. (Color online) Frequency shift as a function of magnetic field applied perpendicular to the crystallographic c axis at different temperatures from 10 to 32 K. The arrow indicates the approximate H_{c2} point for the 24 K field pulse and the linear guides used for determination of the point. Inset: the schematic diagram of the rf sensing coil and sample for the $\mu_0 H \perp c$ configuration.

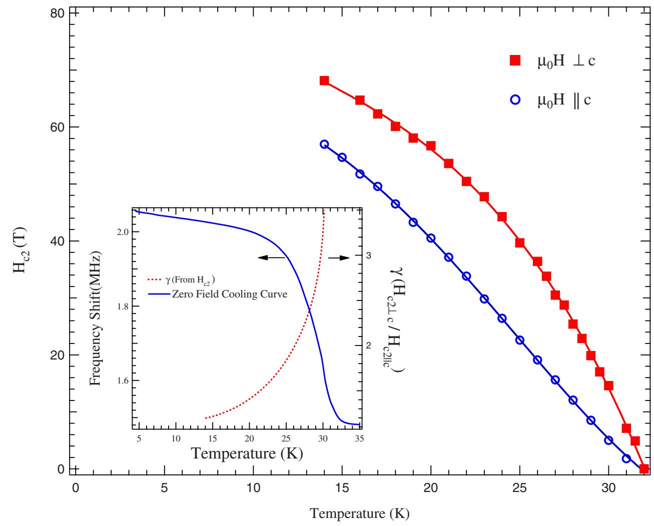

FIG. 3. (Color online) Anisotropic $H_{c2}(T)$ for $(Ba_{0.55}K_{0.45})Fe_2As_2$ single crystals. Circles indicate the $H_{c2}^{\parallel c}$ and squares $H_{c2}^{\perp c}$. Inset: the rf zero-field cooling curve for $(Ba_{0.55}K_{0.45})Fe_2As_2$ single crystals on the left axis and temperature-dependent anisotropy of $H_{c2}: \gamma = H_{c2}^{\perp c}/H_{c2}^{\parallel c}$ as determined from the fits to the H_{c2} data on the right axis.

hibit signs of heating due to eddy currents induced in the sample, caused by the pulsed field. Our results show no hysteresis (hence no measurable heating) during the pulse, indicating a good thermal anchor was established with the bath. To determine the upper critical-field anisotropy the single crystal was measured in two orthogonal sample orientations: with the applied field parallel to the conducting planes [perpendicular to the crystallographic c axis (see inset of Fig. 1)] and with the applied field normal to the conducting planes [parallel to the crystallographic c axis (see inset of Fig. 2)]. Alignment accuracy was within approximately 2°. For applied field perpendicular to the c axis (Fig. 1) the sample was

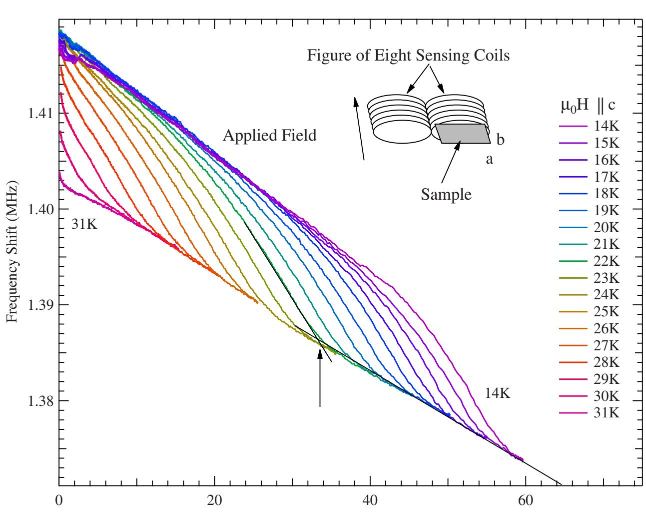

FIG. 2. (Color online) Frequency shift as a function of magnetic field applied along the crystallographic c axis at different temperatures from 14 to 31 K. Inset: the schematic diagram of the rf sensing coil and sample for the $\mu_0 H \| c$ configuration.

placed in a circular detection coil (≈0.8 mm in diameter). The sample and probe were cooled down to liquid-helium temperatures inside of a double-walled nonmetallic cryostat with ≈500 Torr of helium exchange gas. The onset of the superconducting transition is clearly observed (as a resonant frequency shift) near 32 K in zero applied magnetic field (shown in the inset of Fig. 3). In the second orientation (H parallel to the c axis) (Fig. 2) a flux-compensated "figure eight" detection coil set (0.7 mm diameter each) was placed with its symmetry axis parallel to the applied magnetic field. The compensated nature of the detection coil was necessary to minimize induced voltages feeding back into the rf IC. The sample was placed on the top surface of one side of the counterwound coil pair. This configuration has weaker coupling to the sample (<50%), resulting in a smaller but still easily resolvable frequency shift. The temperature was stabilized with a conventional proportional-integral-derivative (PID) temperature controller with an accuracy of $\approx \pm 0.1$ K.

The criterion for determining the point at which the H_{c2} transition occurs requires consistency, and no one method is entirely unambiguous. However, the rf penetration depth technique has demonstrated a high degree of agreement with accepted thermodynamic methods[9] and as a "contactless" method avoids problems with contact lead problems that can be detrimental to transport measurements in pulsed magnetic fields. In addition the rf technique has the advantage of being more sensitive with several-orders-of-magnitude faster time response, hence making it suitable for millisecond duration pulsed magnetic fields (when compared to specific heat, for example). Our method, for determining a consistent H_{c2} point in the Δf vs $\mu_0 H$ data shown in Figs. 1 and 2, is based on identifying the point at which the slope of the rf signal (in the transition region) intercepts the slope of the normal-state background. The justification is simply that the high-field data (above H_{c2}) show a smooth close-to-linear magnetic-field dependence. In this region the rf probe is sensitive to the normal-state magnetoresistance of the sample and detection coil. Below H_{c2} the suppression of the superconducting state with increasing field leads to a field-dependent frequency shift that results in a clear slope change in the data (see Figs. 1 and 2). The H_{c2} value is determined for each subsequent fixed temperature magnetic-field pulse producing the H-T plot (see Fig. 3).

The shapes of the upper critical-field curves for the parallel and perpendicular orientations clearly do not manifest the same temperature dependence. The anisotropy parameter γ (inset of Fig. 3) varies accordingly. Estimates of the anisotropic coherence length at 14 K, based solely on the vortex flux density argument, yield $\xi_{\perp c}(0) \approx 34$ Å (in-plane ξ) and $\xi_{\parallel c}(0) \approx 29$ Å (interlayer ξ) since $\xi_{\perp c} = \sqrt{\phi_0 / 2\pi H_{c2}^{\parallel c}}$ and $\xi_{\parallel c} = \phi_0 / 2\pi \xi_{\perp c} H_{c2}^{\perp c}$.[10] Decoupling of the superconducting layers would occur when $\xi_{\parallel c}(T)$ becomes less than the c-axis spacing, $c \approx 13$ Å for $(Ba_{0.55}K_{0.45})Fe_2As_2$.[8] To estimate the zero-temperature coherence length, we note that $\xi(T)$ is approximated by Ginzburg-Landau (GL) theory, i.e., $\xi(T) \propto \xi(0)/(1-t)^{1/2}$ where $t = T/T_c$. From $\xi_{\parallel c}(14\ \mathrm{K})$ above, $\xi_{\parallel c}(0\ \mathrm{K})$ is about 21.5 Å. Hence although the critical-field anisotropy appears to decrease at lower temperatures, there is no simple argument based on estimates of the temperature-dependent coherence lengths to support a dimensional crossover in the temperature range of this experiment. Increased magnetic-field intensities exceeding 75 T will be required to fully map out the lower-temperature critical-field phase diagram.

The anisotropy of H_{c2} in $(Ba_{0.55}K_{0.45})Fe_2As_2$ is also reported in Ref. 11 by measurement of the electrical resistivity of samples from a different origin. The work presented here differs from Ref. 11 in that a double hump appears in the cooling data in Ref. 11. This artifact may be due to multiple crystalline phases or a doping inconsistency. Our crystals manifest a single and continuous superconducting transition (shown in the inset of Fig. 3).

To summarize, we have measured the anisotropic $H_{c2}(T)$ curves for $(Ba_{0.55}K_{0.45})Fe_2As_2$ down to 14 K and up to 60 T. We find (i) a nonlinear temperature dependence of the H_{c2} anisotropy of $(Ba_{0.55}K_{0.45})Fe_2As_2$, although there is at present no direct evidence for superconducting layer decoupling, and (ii) $H_{c2}(T=0)$ for this compound may easily approach fields of 75 T or above. Higher-field measurements will be required to determine the lower-temperature parts of the full temperature dependence of these remarkably large $H_{c2}(T)$ curves. On the other hand, perhaps even further physical insight will be drawn from careful studies of the angular dependence of $H_{c2}(T)$ for fields below 50 T. Rotations from $H \parallel c$ to $H \parallel [100]$ and $H \parallel [110]$, as well as in-plane rotations studies, should reveal a wealth of details about this modest and temperature-dependent upper critical-field anisotropy.

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