Weak anisotropy of the superconducting upper critical field in Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ single crystals

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We have determined the resistive upper critical field $H_{c2}$ for single crystals of the superconductor Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ using pulsed magnetic fields of up to 60T. A rather high zero-temperature upper critical field of $\mu_0 H_{c2}(0) \approx 47$T is obtained, in spite of the relatively low superconducting transition temperature ($T_c \approx 14$K). Moreover, $H_{c2}$ follows an unusual temperature dependence, becoming almost independent of the magnetic field orientation as the temperature $T \rightarrow 0$. We suggest that the isotropic superconductivity in Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ is a consequence of its three-dimensional Fermi-surface topology. An analogous result was obtained for (Ba,K)Fe$_2$As$_2$, indicating that all layered iron-based superconductors exhibit generic behavior that is significantly different from that of the "high-$T_c$" cuprates.

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The discovery of superconductivity in the iron pnictides LnFeAs(O,F) (where Ln can be La, Ce, Pr, Nd, Sm or Gd) [1, 2, 3, 4, 5] with transition temperatures $T_c$ as high as 55 K has been responsible for something of a resurrection in the study of high temperature superconductivity. Beside the LnFeAs(O,F) series (the so-called “1111s”), other families of the iron-based superconductors have been found, including the “122” materials possessing the ThCr$_2$Si$_2$ structure (e.g., hole- or electron-doped BaFe$_2$As$_2$) [6, 7], the “11-type” LiFeAs family [8, 9] and the “11-type” iron chalcogenides with $\alpha$-PbO structure (e.g., Fe$_{1+x}$Se$_{1-x}$Te) [10, 11]. All of these compounds share a common structural feature, i.e., square planar sheets of Fe, coordinated tetrahedrally by pnictogens or chalcogens. The relatively high superconducting transition temperatures and layered crystal structures of the Fe-based superconductors initially suggested strong analogies with the cuprates. However, in this letter we report pulsed-field magnetoresistance measurements for single crystals of Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ that show that its upper critical field attains a value of 47T as temperature $T \rightarrow 0$ that is almost independent of field direction. This suggests that the electronic properties of Fe$_{1+x}$Te$_{1-x}$ superconductors are rather isotropic (i.e., three dimensional), in complete contrast to those of the quasi-two-dimensional cuprates. A similar effect was found in (Ba,K)Fe$_2$As$_2$ [12] and other 122-type systems [13, 14, 15], indicating this may be a general feature of all iron pnictides.

Large single crystals of Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ were grown by a self-flux method. The starting composition was Fe(Te$_{0.6}$Se$_{0.4}$)$_{0.85}$. The mixtures of Fe, and (Te,Se) were ground thoroughly and sealed in an evacuated quartz tube. The tube was heated to 920°C and cooled slowly to grow large single crystals. The crystals obtained were checked by X-ray diffraction (XRD); their composition was analyzed using a scanning electron microscope (Hitachi S4000) equipped with an Energy Dispersive X-Ray Spectrometer (EDXS). Longitudinal resistivity was measured using a typical four-contact method in pulsed fields of up to 60T at the National High Magnetic Field Laboratory, Los Alamos [12]. In order to minimize inductive self-heating caused by the pulsed magnetic field, small crystals with typical sizes $2 \times 0.5 \times 0.1$ mm$^3$ were cleaved off along the c-direction from the as-grown samples. Data were recorded using a 10 MHz digitizer and 100 kHz alternating current, and analyzed using a custom low-noise digital lock-in technique [12]. Care was taken to ensure that neither the current nor the field pulse caused significant heating. The temperature dependence of the resistivity at zero field was measured with a Lakeshore resistance bridge. Complementary magnetization data $M(T)$ were measured using a Quantum Design SQUID magnetometer.

Figure 1 presents the temperature dependence of the in-plane electrical resistivity $\rho_{ab}(T)$ for Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ at zero field. As reported in the literature [16, 17], Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ exhibits a resistivity that increases with decreasing temperature. Nevertheless, it undergoes a relatively sharp superconducting transition at $T_c = 14 \pm 0.3$K. Bulk superconductivity is confirmed by the temperature dependence of the dc magnetic susceptibility, as plotted in the inset of Fig.1.

The field dependent electrical resistivity, $\rho(H)$, at various temperatures is shown in Fig. 2(a) and Fig. 2(b) for magnetic fields applied parallel $(H \parallel c)$ and perpendicular to $(H \perp c)$ the c-axis, respectively. For consistency, only data collected during the down-sweep of the magnet are shown. The superconducting to normal transition is visible as a sharp rise in $\rho$: inside the superconducting
state, an apparent finite $\rho$ is observed for $\mathbf{H} \parallel \mathbf{c}$, but not for $\mathbf{H} \perp \mathbf{c}$. The former behavior is likely to be due to dissipation associated with thermally-activated flux motion [18]. Nevertheless, it is obvious that at the same temperature, superconductivity is suppressed by a large magnetic field [12, 19], suggesting that this behavior might be a more general phenomenon that is not primarily associated with excess Fe [17].

The temperature dependence of the upper critical field $H_{c2}(0)$ of Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$, determined from the mid-point of the sharp resistive superconducting transitions, as shown in Fig. 2, is plotted in Fig. 4 for magnetic field parallel and perpendicular to the $c$-axis. The two crystals (samples A and B, with $T_c = 14 \pm 0.3K$) exhibit an almost identical behavior of $H_{c2}$, indicating good sample reproducibility. The most remarkable aspect of Fig. 4 is the fact that the upper critical fields of Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$ for the two field orientations merge together as $T \to 0$ at $\mu_0 H_{c2} \approx 47$ T. This $H_{c2}(0)$ is consistent with the value determined for the polycrystalline sample [20].

The anisotropy coefficient $\gamma(T)$, determined from

$$\gamma(T) = H_{c2}^{\parallel}/H_{c2}^{\perp}$$

decreases monotonically from 2 near $T = T_c$ to about 0.95 at $T = 0$ (see the lower inset of Fig.4). Similar isotropic behavior of the upper critical field has also been observed in the 122-series of Fe-based superconductors [12] [13] [14] [15]. All these results indicate that nearly isotropic superconductivity might be a general, but very unique feature, of the iron-based superconductors.

The anisotropy of the upper critical field is usually determined by the underlying electronic bandstructure. In the layered cuprates and organic superconductors, the Fermi surfaces are rather two-dimensional [21] [22]. As a result, there is considerable anisotropy; the upper critical field of these materials is large for in-plane fields, being determined by spin mechanisms such as the Pauli paramagnetic limit, but generally much smaller and restricted by orbital mechanisms for other field orientations [21] [23]. However, the experiments in this paper show that this is not the case for Fe$_{1.11}$Te$_{0.6}$Se$_{0.4}$; its upper critical field $H_{c2}$ at low temperature displays only a very weak de-
The data sets indicate good sample reproducibility. These data sets indicate good sample reproducibility. The inset plots the superconducting transitions in detail.

In summary, we have determined the resistive upper critical field for sample A (main plot) and sample B (upper inset) where the solid and open symbols represent \( H \parallel c \) and \( H \perp c \), respectively. These data sets indicate good sample reproducibility of \( H_{c2}(T_c) \). The lower inset plots the anisotropic coefficient \( \gamma (= H_{c2}^+ / H_{c2}^-) \) as a function of temperature for sample B.

Although the low-temperature upper critical field is rather isotropic, the initial slope of \( H_{c2} \) near \( T_c \) does show some dependence on the field orientation (Fig. 4); similar behavior in the 122 compounds has been attributed to two-band superconductivity \cite{21, 22, 23, 24, 25, 26, 27}. In our resistive critical field data, \( dH_{c2}/dT(T = T_c) \) is about 8.90 T/K for \( H \perp c \) and 3.82 T/K for \( H \parallel c \), respectively. These are close to the values observed for Fe\(_{11.1}\)Te\(_0.6\)Se\(_{0.4}\) in dc field measurements \cite{16, 20}. Upon cooling down, \( H_{c2}(T) \) for \( H \perp c \) starts to bend down, resulting in a significantly lower zero temperature upper critical field compared to typical extrapolation methods. For example, the upper critical field at \( T = 0 \) determined by the Werthamer-Helfand-Hohenberg (WHH) theory \cite{29} yields a value of about 87 T for \( H \perp c \), \( T_c \approx 2.65 \text{ nm} \) as 2.65 nm.
dimensional nature of their Fermi-surface topology. This is in great contrast to the cases of high $T_c$ cuprates and organic superconductors which possess highly anisotropic upper critical fields due to their quasi-two-dimensional band structure. As mentioned in Ref. [30], our findings of isotropic superconductivity together with a rather high upper critical field suggest that the iron-based superconductors are very promising materials for future applications, in particular if $T_c$ could be further enhanced above nitrogen temperature.

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