Upper Critical Field of the Stoichiometric Fe-based Superconductor LiFeAs

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Abstract.
We have investigated the upper critical field $B_{c2}$ of the stoichiometric Fe-based superconductor LiFeAs, via the magnetic torque measurements in dc-magnetic fields up to 35 T and at temperatures down to 0.3 K. $B_{c2}$ at 0.3 K is obtained to be 26.4 T and 15.5 T for the applied field $B_a \parallel ab$ and $B_c \parallel c$, respectively, indicating nearly-isotropic superconductivity. The detailed analyses show that, while $B_{c2}$ for $B_a \parallel c$ is limited by the orbital effect, the spin-paramagnetic effect is evident in the temperature dependence of $B_{c2}$ for $B_a \parallel ab$.

1. Introduction
The discovery of the Fe-based superconductors (SCs) [1] has triggered intensive research efforts, not only due to the high transition temperature $T_c$ (up to 54 - 56 K in some compounds), but the possible unconventional pairing state, a variety of the FeAs layered crystal structures, etc. [2].

As to the upper critical field $B_{c2}$ of Fe-based SCs, thus far, there are few reports on the systematic investigations to address the issue of the pair-breaking mechanism. This is mainly due to the fact that high-$T_c$ SCs including Fe-based systems generally have extremely high $B_{c2}$. In many cases, accordingly, the low temperature behavior is extrapolated from the high temperature data around $T_c$. However, such an extrapolation, generally assuming an orbitally limited case [3], could lead to misleading conclusions for the Fe-based systems where a spin-paramagnetic effect [4] may play an important role. For better understanding of the pair-breaking mechanism of the high-$T_c$ superconductivity, therefore, it is of importance to determine $B_{c2}$ experimentally in a wide temperature range from $T_c$ down to $T \rightarrow 0$.

Recently, we have performed magnetic torque measurements of the stoichiometric Fe-based superconductor LiFeAs [5, 6] using a single crystal with $T_c \sim 16$ K, and have first reported on the whole temperature dependence of $B_{c2}$ and its anisotropy [8]. Here, we present more detailed data which supplement the results of Ref. [8].
2. Experimental Details
LiFeAs single crystals with the residual resistivity ratio of $\sim 45$ were grown by a self-flux method as described in Ref [7]. The samples were carefully handled so as not to expose them to air directly. The bulk superconductivity was confirmed from the clear diamagnetic transition of the dc magnetic susceptibility below $T_c \sim 16$ K [8]. The magnetic torque measurements were carried out using a static resistive magnet in fields up to 35 T and at temperatures down to 0.3 K, by means of a piezoresistive microcantilever. The specific heat $C$ was measured in a physical property measurement system (PPMS; Quantum Design). The nuclear magnetic resonance (NMR) spin-lattice relaxation time $T_1$ was measured for the $^{75}$As NMR lines. The crystalline axes were aligned within $1^\circ$ by using a two-axis goniometric stage during the NMR experiment.

3. Results and Discussions
Figures 1(a) and (b) show the torque signals in LiFeAs as a function of the applied field $B_a$ for $B_a \parallel ab$ and $B_a \parallel c$, respectively, at several temperatures down to 0.3 K. As indicated by arrows, highly hysteretic behavior, which is more significant at low temperatures, is observed between the field - up and - down sweeps. The hysteresis of the torque response, the irreversible curve, appears in the superconducting mixed state when the pinning force is strong enough to trap the flux lines. Note that the shapes of the small loops near $B_{c2}$ for $B_a \parallel ab$ and $B_a \parallel c$ are

![Figure 1](image-url). (Color online) Magnetic torque signals vs $B_a$ of LiFeAs at several temperatures down to 0.3 K for (a) $B_a \parallel ab$ and (b) $B_a \parallel c$. The solid curves represent the data obtained in field-up and -down sweeps between $B_a=0$ and above $B_{c2}$, where the arrows in (a) and (b) indicate the sweep directions. Small-loop measurements around $B_{c2}$, as shown by the dashed curves at $T=0.3$ K, were performed at each temperature. (c) $C/T$ vs $T$ and (d) $1/(T_1 T)$ vs $T$ of LiFeAs in several fields for $B_a \parallel c$. The arrows in (c) and (d) indicate $B_{c2}$ at each field.
Dashed curves indicate fits to the data based on the WHH theory (see text). (b) Scaled \( \frac{B_{c2}}{T_c} \) vs \( \frac{T}{T_c} \) of LiFeAs. For comparison, data obtained from TDO [14] and \( \rho \) [15, 16] are also shown.

Figure 2(a) displays \( B_{c2} \) vs \( T \) of LiFeAs for \( B_a \parallel ab \) and \( B_a \parallel c \), deduced from the magnetic torque, \( C \), \( ^{75} \text{As-NMR} \), and \( \chi_{ac} \) measurements. The dashed curves contain the spin-paramagnetic and orbital pair-breaking effects [3]. From the best fits, the Maki parameter \( \alpha \) and spin-orbit scattering parameter \( \lambda_{so} \) are obtained to be \( (\alpha, \lambda_{so}) = (2.30, 0.51) \) for \( B_a \parallel ab \) and \( (0.75, \infty) \) for \( B_a \parallel c \), where \( T_c = 15.5 \text{ K} \) is fixed. The orbital critical fields \( B_{c2}^* \) at \( T = 0 \) are estimated to be 47.2 and 15.3 T for \( B_a \parallel ab \) and \( B_a \parallel c \), respectively, from \( B_{c2}^*(0) = 0.69 T_c \frac{dH_{c2}}{dT}|_{T=T_c} \) [3]. The Pauli-Clogston paramagnetic limit \( B_{po} = 1.84 T_c \) [4] is 28.5 T.

It is interesting to note that \( B_{c2}^*(0) < B_{po} < B_{c2}^{*ab}(0) \). This relation results in the orbitally limited \( B_{c2} \) for \( B_a \parallel c \), as indicated by \( \lambda_{so} = \infty \), and strongly spin-paramagnetically limited \( B_{c2} \) for \( B_a \parallel ab \), as indicated by that \( B_{c2}^{*ab}(0.3 \text{ K}) = 26.4 \text{ T} \ll B_{c2}^{*ab}(0) \), and effectively reduces the \( B_{c2} \) anisotropy at low temperatures. A similar trend that the weaker orbital effect for \( B_a \parallel ab \) is partly compensated for by the spin-paramagnetic effect to yield a reduced anisotropy is clearly seen in a stoichiometric superconductor KFe2As2 with low \( T_c \) of 2.8 K [10], as well as in the...
high-$T_c$ systems such as “122”-type (Ba,K)Fe$_2$As$_2$ [11] and “11”-type FeSe [12].

As temperature decreases, the anisotropy parameter $\Gamma$, defined as $\Gamma = B_{c2}^{ab} / B_{c2}^c$, decreases from 3.1 at $T_c$ (from the WHH fit) to 1.7 at 0.3 K. The low-temperature variation of $\Gamma$ is similar to those observed in the “122” and “11” systems [11, 12], but the value of $\Gamma$ in LiFeAs is slightly larger. The slightly larger $\Gamma$ of LiFeAs than those of the “122” and “11” systems might be due to the more two-dimensionality of LiFeAs [13].

Soon after the publication of our study [8], three groups independently reported on the full $B_{c2} - T$ phase diagram of LiFeAs using different techniques: tunnel diode resonator (TDO) [14], electrical resistivity (pulse magnet) [15], and (static magnet) [16]. In Fig. 2(b), we show a comparison of the scaled $B_{c2}/T_c$ vs $T/T_c$. Note that our results are qualitatively consistent with others in terms of that the pair-breaking for $B_a \parallel c$ is mostly caused by the orbital effect while a spin-paramagnetic effect plays an important role for $B_a \parallel ab$. The recognizable discrepancies may be explained by a combination of several factors such as the manner to define the $B_{c2}$ value, the sample quality which affects $B_{c2}$, and so on.

4. Conclusions
To conclude, we have performed high-field magnetic torque measurements of the LiFeAs single crystal, and have determined the full temperature dependence of $B_{c2}$ down to 0.3 K. The detailed analyses show that, while $B_{c2}$ for $B_a \parallel c$ is limited by the orbital effect, the weaker orbital effect for $B_a \parallel ab$ due to the mass anisotropy is partly compensated for by the spin-paramagnetic effect leading to a reduced anisotropy at low temperatures.

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References
[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] For recent reviews, see, Ishida K, Nakai Y and Hosono H 2009 J. Phys. Soc. Jpn. 78 062001; Johnston D C 2010 Adv. Phys. 59, 803, and references therein.
[3] Werthamer N R, Helfand E and Hohenberg P C 1966 Phys. Rev. 147 295
[4] Chandrasekhar B S 1962 Appl. Phys. Lett. 1 7; Clogston A M 1962 Phys. Rev. Lett. 9 266
[5] Wang X C, Liu Q Q, Lv Y X, Gao W B, Yang L X, Yu R C, Li F Y and Jin C Q 2008 Solid State Commun. 148 538
[6] Tapp J H, Tang Z, Lv B, Samal K, Lorenz B, Chu P C W and Guloy A M 2008 Phys. Rev. B 79 060505
[7] Imai Y, H. Takahashi, K. Kitagawa, K. Matsubayashi, N. Nakai, Y. Nagai, Y. Uwatoko, M. Machida, and A. Maeda 2011 J. Phys. Soc. Jpn. 80 013704
[8] Kurita N, Kitagawa K, Matsubayashi K, Kismahardja A, Choi E S, Brooks J S, Uwatoko Y, Uji S and Terashima T 2011 J. Phys. Soc. Jpn. 80 013706
[9] Matsubayashi K and Kitagawa K, private communications.
[10] Terashima T, Kimata M, Satsukawa H, Harada A, Hazama K, Uji S, Harima H, Chen G F, Luo J L and Wang N L 2009 J. Phys. Soc. Jpn. 78 063702
[11] Yuan H Q, Singleton J, Balakirev F F, Baily S A, Chen G F, Luo J L and Wang N L 2009 Nature 457 565
[12] Fang M, Yang J, Balakirev F F, Kohama Y, Singleton J, Qian B, Mao Z Q, Wang H and Yuan H Q 2010 Phys. Rev. B 81 020509(R).
[13] Singh D J 2008 Phys. Rev. B 78 094511.
[14] Cho K, Kim H, Tanatar M A, Song Y J, Kwon Y S, Coniglio W A, Agosta C C, Gurevich A and Prozorov R 2011 Phys. Rev. B 83 060502(R)
[15] Zhang J L, Jiao L, Balakirev F F, Wang X C, Jin C Q and Yuan H Q 2011 Phys. Rev. B 83 174506
[16] Khim S, Lee B, Kim J W, Choi E S, Stewart G R and Kim K H 2011 Phys. Rev. B 84 104502