Two first-order phase transitions demarcating the peak-effect region in iron-based superconductors

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We report magnetic torque measurements on iron-pnictide superconductors Ba₁₋ₓKₓFe₂As₂ (x = 0.69 and 0.76) up to an applied field of B₀ = 45 T. The peak effect is observed below the irreversibility field and is enhanced as the field is tilted from the c axis. For some tilted field directions, the hysteresis loop exhibits very sharp features indicative of two first-order phase transitions that demarcate the peak-effect region. We construct and discuss the applied-magnetic-field (B₀)–temperature (T) phase diagram.

The critical current in type-II superconductors often shows an anomalous peak just before it becomes zero at the upper critical field Bc₂. This ‘peak effect’ has been known since the early 1960’s and has attracted continuing attention [1, 2]. Its mechanism however still remains unresolved. Not only will its elucidation deepen our understanding of the vortex-matter physics, it may also be of technological importance: it might open a new avenue to improve or tailor the critical current.

One of plausible explanations associates the peak effect with an order–disorder transition of the vortex lattice [3, 4]. The applied-magnetic-field (B₀)–temperature (T) phase diagram of ideal type-II superconductors consists of the Meissner and mixed states. However, in real materials, the perfect Abrikosov vortex lattice does not exist, and the mixed state is subdivided into different vortex states. A quasi-long-range-ordered Bragg glass occupies a low-T low-B₀ part of the mixed state in weak-pinning superconductors [5, 6]. The Bragg glass melts into a vortex liquid as the temperature increases. On the other hand, increasing magnetic field is equivalent to increasing pinning strength. As the field is increased at low temperatures, the Bragg glass thus disorders at a certain field to better adapt to the random pinning environment, resulting in a larger critical current. Although the nature of the disordered phase is still controversial, it is widely believed that this order-disorder transition underlies the peak effect [3, 4]. Experimental evidence has accumulated, especially in low-Tc materials: Magnetic measurements showed anomalous field- or temperature-history dependence in the peak effect region [7–9]. The coexistence of two phases with differing critical currents in the peak-effect region were directly seen by scanning Hall-probe microscopy [10]. Small angle neutron scattering revealed disordering of the vortex lattice near the peak-effect region [11, 12].

In this article, we report magnetic torque measurements on iron-pnictide superconductors Ba₁₋ₓKₓFe₂As₂.

FIG. 1. Hysteresis loops of Ba₀.₃₁K₀.₆₉Fe₂As₂ at T = 0.4 K. Magnetic torque divided by applied field is shown as a function of applied field for various field angles θ. The solid and broken curves show increasing- and decreasing-field ones, respectively, as indicated by the arrows.

Unlike the outline above, we find two first-order phase transitions that demarcate the peak-effect region for some field directions. We construct and discuss the B₀-T phase diagram.

Single crystals of Ba₁₋ₓKₓFe₂As₂ (x = 0.69 and 0.76) were synthesized by a KAs self-flux method [14, 15]. Small pieces with typical dimensions of (50–100 µm)²×(a few tens of µm) were prepared by cleaving crystals along <100> axes. The 45-T hybrid magnet or a 35-T resistive magnet was used with a ³He refrigerator at the NHMFL in Tallahassee. The magnetic torque τ = M × B₀ was measured with a piezo-resistive microcantilever [16]. The angle θ of the applied field B₀ was measured from the c axis.

Figure 1 shows the magnetic torque divided by the
The peak effect hence manifests itself as enhancement of the critical current, or the pinning force, at that field. Decreasing-field curves at a given field is a measure of how much pinning is weaker above the transition field. Therefore the anomalies at \( B_1^- \) and \( B_2^- \) are not artifacts caused by field sweeping, we have taken relaxation data at fields indicated by hollow circles. The inset of (a) shows a full hysteresis curve, while the main panel shows a plot of local fields inside a slab of thickness \( d \) parallel to the surface.

FIG. 2. Hysteresis loops and history dependence for Ba\(_{0.24}\)K\(_{0.76}\)Fe\(_2\)As\(_2\) at \( \theta = 82^\circ \) and \( T = 0.45 \) K. (a) Enlarged view of the peak effect region. The field sweep direction is indicated by arrows. The solid line was obtained from a continuous field sweep, while circles by stopping field at some value and waiting for about one minute to see effects of relaxation. See text for the definitions of the transition fields \( B_1^+(-) \) and \( B_2^+(-) \). The full hysteresis curve is shown in the inset. (b) Minor hysteresis loops showing history effects. Solid curves are obtained by increasing field from 0 T to some field and then decreasing field, while broken ones by decreasing field from 33 T to some field and then increasing field, as indicated by arrows. The inset is an enlarged view of a region near the irreversibility field and upper critical field.

axis towards the \( ab \) plane with increasing \( \theta \). The peak becomes asymmetric at large angles: an increasing- and a decreasing-field curve peak at markedly different fields. An increasing-field curve shows a sharper slope on the high-field side of the peak while a decreasing-field one shows a sharper slope on the low-field side.

FIG. 3. Explanation of the asymmetric hysteresis loops within the Bean model [13], which assumes that the field gradient develops in a superconducting sample according to \( dB/dx = \mu_0 J_c \) where \( J_c \) is a critical current density. Shown are plots of local fields inside a slab of thickness \( d \) with applied fields parallel to the surface. \( J_c = J_0, J_1, \) and \( J_2 \) for \( B_a < B_1^+ \), \( B_1^+ < B_a < B_2^+ \), and \( B_2^+ < B_a \), respectively, and \( J_1 \gg J_0, J_2 \). When \( B_1^+ \) is crossed from below to above, the field gradient inside the slab changes only gradually from the surface, and hence the width of the transition region \( \Delta B_1^+ \) is large. On the other hand, when \( B_2^+ \) is crossed, the critical current collapses and can no longer sustain the existing field gradient, giving rise to a quick change in the magnetization. The transition width \( \Delta B_2^+ \) is much smaller.
FIG. 4. (a) Hysteresis loops of $\text{Ba}_{0.24}\text{K}_{0.76}\text{Fe}_2\text{As}_2$ for various temperatures. The irreversibility field $B_{irr}$ where the increasing- and decreasing-field branches diverge and upper critical field $B_{c2}$ where the superconducting diamagnetism appears are marked for the $T = 12$ K curve. (b) $B_a$-$T$ phase diagram determined from (a).

is crossed from below to above or $B_{c2}^−$ from above to below, the sample enters a strongly-pinned state. Since the field gradient built in a weakly-pinned state can be sustained by a large critical current in the strongly-pinned state, the change in the field gradient occurs only gradually from the surface. Therefore only a bend in the $\tau/B$ curve is observed at $B_{c1}^+$ or $B_{c2}^-$. On the other hand, when $B_{c2}^+$ is crossed from below to above or $B_{c1}^-$ from above to below, the sample enters a weakly-pinned state from the strongly-pinned one. Hence the field gradient built in the strongly-pinned state becomes no longer sustainable, and the field gradient quickly changes throughout the sample so that it becomes small enough to be sustained by a small critical current in the weakly-pinned state. This gives rise to a sudden change in the sample magnetization. (Quantitative considerations of the transition widths are given below.)

Figure 2(b) shows various minor hysteresis loops. The curve branching off from the increasing-field curve at a field below $B_{c1}^+$ undershoot the decreasing-field curve of the full loop, while those branching off at fields above $B_{c2}^+$ overshoot. Also, curves branching off from the decreasing-field curve at low fields go slightly below the increasing-field curve of the full loop. These observations indicate complicated phase coexistence due to the first-order phase transitions.

Another noteworthy feature in Fig. 2(b) is that the branched-off curves traverse a large field difference to approach the opposite side of the full loop. The curve branching off from the increasing-field curve at $B_a = 23.4$ T just below $B_{c2}^+$ does not reach the decreasing-field curve until 19.5 T, for example. Within the Bean model, a curve branching off from the increasing-field (decreasing-field) curve of the full hysteresis loop at $B_a = B_o - 2\Delta B_1(B_o + 2\Delta B_1)$, where $\Delta B_1 = \mu_0J_1d/2$ is the field difference between the sample surface and center in the strongly-pinned state (Fig. 3). Let us assume $J_1 = 10^8$ A/cm$^2$ as $J_c$ of this magnitude has been observed at low fields in doped $\text{BaFe}_2\text{As}_2$ [17–19]. Since the applied field is roughly parallel to the surface, we might take the sample thickness as $d$: then, $d \sim 0.02$ mm. This gives $2\Delta B_1 \sim 0.02$ T, too small to explain the observation. It may be more appropriate to decompose the magnetization and applied field into the $c$-axis and $ab$-plane components. In the case of the above-mentioned curve, the $c$-axis component of the applied field ($B_a \cos \theta$) changes from 3.3 to 2.7 T. The torque is given by $M_eB_a^c - M_{ab}B_a^c$ and is dominated by the first term. Since the $c$-axis magnetization $M_e$ is caused by the shielding of the $c$-axis component of the field $B_a^c$, the relevant dimension now is the sample length: then $d \sim 0.1$ mm. This gives $2\Delta B_1 \sim 0.1$ T for the $c$-axis component, which does not seem sufficient to explain the observation. We also note the curves branching off at 23.8 T, which is definitely above $B_{c2}^+$ and hence the sample is in the weak-pinning state. Within the Bean model the curve is expected to approach the decreasing-field curve much more quickly, but it actually goes nearly parallel to the above-discussed curve branching off at $B_a = 23.4$ T. Clearly, the behavior of minor hysteresis loops cannot fully be understood within the Bean model, and it seems necessary to involve complex phase coexistence.

Figure 4(a) shows temperature variation of magnetic torque curves measured on another sample. As the temperature is raised, the anomalies at $B_{c1}^-$ and $B_{c2}^+$ becomes less sharp, and the increasing- and decreasing-curves become more symmetric. The irreversibility field clearly separates from the upper critical field as indicated for the $T = 12$ K curve. Figure 4(b) shows the determined $B_a$-$T$ phase diagram.
If we assume that the phase below $B_1$ is the Bragg glass, the present phase diagram can be interpreted as follows: The phase between $B_1$ and $B_2$ is a disordered solid phase, which may be a vortex glass [20] or multidomain glass [21]. The phase above $B_2$ is a vortex liquid, and the irreversibility line is a crossover line separating a pinned and an unpinned liquid. This interpretation is similar to a proposal in [3, 21]. We, however, note the following: those previous works were based on the observation of a single peak of $J_\parallel$ in the peak effect region and associated it with the boundary between the disordered solid and liquid phases. A very recent small angle neutron scattering study on vanadium, however, claims that the peak effect lies at higher fields and temperatures than the order-disorder transition [22].

On the other hand, recent STM studies of the vortex lattice in Co$_{0.0075}$NbSe$_2$ indicate that disordering of the Bragg glass occurs via two phase transitions, i.e., from the ordered state through an orientational glass to the amorphous vortex glass [23, 24]. It is noteworthy that superheating and supercooling effects are observed across either transition. Two-step disordering has also been reported in a numerical study [25]. Our $B_1$ and $B_2$ phase transitions might correspond to those two transitions. It is however to be noted that those studies are for $B \parallel c$. In the present case, the field is tilted from the $c$ axis. It may be necessary to consider the two components of $J_\parallel$, i.e., $J_\parallel \parallel c$ and $J_\parallel \perp c$, to explain the existence of the two transitions.

Two more features in Fig 4(b) are noteworthy: The two transition fields $B_2$ and $B_{c2}$ are separate at the lowest temperature of $T = 0.47$ K, suggesting the existence of a quantum vortex liquid at $T = 0$, although it is pinned. Secondly, although the irreversibility field $B_{irr}$ is distinct from the upper critical field $B_2$ at high temperatures, they coincide (within experimental accuracy) as $T$ approaches zero. This may have implications for an ongoing debate about the exact location of the upper critical field in high-$T_c$ cuprates [26–28].

In summary, we have performed magnetic torque measurements on single crystals of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.69$ and 0.76) and found for inclined filed directions two successive first-order phase transitions defining the peak effect region. We have discussed possible origins of the two phase transitions, but further studies are necessary to conclude.

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[1] T. G. Berlincourt, R. R. Hake, and D. H. Leslie, Phys. Rev. Lett. 6, 671 (1961).
[2] A. B. Pippard, Phil. Mag. 19, 217 (1969).
[3] S. Banerjee, A. Grover, M. Higgins, G. I. Menon, P. Mishra, D. Pal, S. Ramakrishnan, T. C. Rao, G. Ravikumar, V. Sahni, S. Sarkar, and C. Tomy, Physica C: Superconductivity 355, 39 (2001).
[4] T. Giamarchi and B. S., in High Magnetic Fields: Applications in Condensed Matter Physics and Spectroscopy, edited by C. Berthier, L. P. Levy, and G. Martinez (Springer-Verlag, Berlin, 2002) pp. 314 – 360.
[5] T. Giamarchi and P. Le Doussal, Phys. Rev. B 52, 1242 (1995).
[6] T. Klein, I. Joumard, S. Blanchard, J. Marcus, R. Cubitt, T. Giamarchi, and P. Le Doussal, Nature 413, 404 (2001).
[7] S. B. Roy, P. Chaddah, and S. Chaudhary, Phys. Rev. B 62, 9191 (2000).
[8] G. Ravikumar, P. K. Mishra, V. C. Sahni, S. S. Banerjee, A. K. Grover, S. Ramakrishnan, P. L. Gammel, D. J. Bishop, E. Bucher, M. J. Higgins, and S. Bhattacharya, Phys. Rev. B 61, 12490 (2000).
[9] M. Angst, R. Puzniak, A. Wisniewski, J. Jun, S. M. Kazakov, and J. Karpinski, Phys. Rev. B 67, 012502 (2003).
[10] M. Marchevsky, M. J. Higgins, and S. Bhattacharya, Nature 409, 591 (2001).
[11] P. L. Gammel, U. Yaron, A. P. Ramirez, D. J. Bishop, A. M. Chang, R. Ruel, L. N. Pfeiffer, E. Bucher, G. D’Anna, D. A. Huse, K. Mortensen, M. R. Eskildsen, and P. H. Kes, Phys. Rev. Lett. 80, 833 (1998).
[12] X. S. Ling, S. R. Park, B. A. McClain, S. M. Choi, D. C. Dender, and J. W. Lynn, Phys. Rev. Lett. 86, 712 (2001).
[13] C. P. BEAN, Rev. Mod. Phys. 36, 31 (1964).
[14] K. Kihou, T. Saito, S. Ishida, M. Nakajima, Y. Tomioka, H. Fukazawa, Y. Kohori, T. Ito, S. Uchida, A. Iyo, C. H. Lee, and H. Eisaki, J. Phys. Soc. Jpn. 79, 124713 (2010).
[15] K. Kihou, T. Saito, K. Fujita, S. Ishida, M. Nakajima, K. Horigane, H. Fukazawa, Y. Kohori, S. Uchida, J. Akimitsu, A. Iyo, C. H. Lee, and H. Eisaki, J. Phys. Soc. Jpn. 85, 034718 (2016).
[16] E. Ohmichi and T. Osada, Rev. Sci. Instrum. 73, 3022 (2002).
[17] M. A. Tanatar, N. Ni, C. Martin, R. T. Gordon, H. Kim, V. G. Kogan, G. D. Samolyuk, S. L. Bud’ko, P. C. Canfield, and R. Prozorov, Phys. Rev. B 79, 094507 (2009).
[18] L. Fang, Y. Jia, A. Schluetter, A. Kayani, Z. L. Xiao, H. Claus, U. Welp, A. E. Koshelev, G. W. Crabtree, and W.-K. Kwok, Phys. Rev. B 84, 140504 (2011).
[19] S. Demirdi, Y. Fasano, S. Kasahara, T. Terashima, T. Shibauri, Y. Matsuda, M. Konczykowski, H. Pastoriza, and C. J. van der Beek, Phys. Rev. B 87, 094506 (2013).
[20] T. Giamarchi and P. Le Doussal, Phys. Rev. B 55, 6577
(1997).

[21] G. I. Menon, Phys. Rev. B 65, 104527 (2002).

[22] R. Toft-Petersen, A. B. Abrahamsen, S. Balog, L. Porcar, and M. Laver, Nature Communications 9, 901 (2018).

[23] S. Chandra Ganguli, H. Singh, G. Saraswat, R. Ganguly, V. Bagwe, P. Shirage, A. Thamizhavel, and P. Raychaudhuri, Scientific Reports 5, 10613 (2015).

[24] S. C. Ganguli, H. Singh, I. Roy, V. Bagwe, D. Bala, A. Thamizhavel, and P. Raychaudhuri, Phys. Rev. B 93, 144503 (2016).

[25] C. Dasgupta and O. T. Valls, Phys. Rev. Lett. 91, 127002 (2003).

[26] G. Grissonnanche, O. Cyr-Choinière, F. Laliberté, S. R. de Cotret, A. Juneau-Fecteau, S. Dufour-Beauséjour, M.-E. Delage, D. LeBœuf, J. Chang, B. Ramshaw, D. Bonn, W. Hardy, S. A. R. Liang and, N. Hussey, B. Vignolle, C. Proust, M. Sutherland, S. Krämer, J.-H. Park, D. Graf, N. Doiron-Leyraud, and L. Taillefer, Nat. Commun. 5, 3280 (2014).

[27] J. F. Yu, B. J. Ramshaw, I. Kokanović, K. A. Modic, N. Harrison, J. Day, R. Liang, W. N. Hardy, D. A. Bonn, A. McCollam, S. R. Julian, and J. R. Cooper, Phys. Rev. B 92, 180509 (2015).

[28] F. Yu, M. Hirschberger, T. Loew, G. Li, B. J. Lawson, T. Asaba, J. B. Kemper, T. Liang, J. Porras, G. S. Boebinger, J. Singleton, B. Keimer, L. Li, and N. P. Ong, Proc. Nat. Acad. Sci. U. S. A. 113, 12667 (2016).