Multiple Magnetization Peaks and New Type of Vortex Phase Transitions in \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \)

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Magnetization and its relaxation have been measured on \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \) single crystals with \( T_c = 39 \text{ K} \). The magnetization hysteresis loops (MHLs) exhibit flux jumps in the low temperature region, and a second peak-effect in the intermediate temperature region, especially when the field sweeping rate is low. Interestingly a third magnetization peak can be easily observed on the MHLs in the high temperature region. Further analysis find that the first magnetization peak is very sharp, which is associated with the strong vortex pinning. However the first dip of the MHL corresponds to a moderate relaxation rate, then a second peak appears accompanied by a vanishing vortex motion. Finally a third magnetization peak emerges and the vortex motion becomes drastic beyond this threshold. The novel features accompanying the second magnetization peak suggest a new type of vortex phase transition.

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Investigating the vortex phase diagram and its new feature in unconventional superconductors is very important for both of the fundamental sciences and potential applications. In cuprate superconductors, the vortex physics was boosted greatly with the discovery of many interesting vortex phases and the transitions between them.\(^1\)\(^-\)\(^8\). Magnetic hysteresis was used as an important tool to probe the vortex pinning mechanism, vortex dynamics and vortex phase transitions.\(^1\)\(^-\)\(^8\)\(^9\). The second-peak of magnetization (namely, the increase of magnetization with magnetic field) was observed in \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8-\delta \) (Bi-2212) and was understood as a disorder induced first order transition (transition from a quasi-ordered Bragg glass to disordered vortex glass).\(^1\)\(^0\)\(^1\)\(^1\). While in a less anisotropic material, like \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) (YBCO), the broad second-peak of magnetization was called the fish-tail effect, which was explained as due to the crossover from an elastic to plastic pinning regimes.\(^1\)\(^2\) Therefore the second-peak effect, in either the form in YBCO or in Bi-2212, occurs quite frequently in cuprate superconductors.

In 2008 the discovery of superconductivity above 50 K in iron pnictides has added a new member in the family of unconventional high temperature superconductors.\(^1\)\(^3\). The vortex matter and phase diagram in this new kind materials were investigated immediately by a variety of experiments.\(^1\)\(^4\)\(^-\)\(^2\)\(^0\). For example, the magnetic hysteresis loops have been investigated and the second-peak (or fish-tail) effect was also observed in the 122, 1111 and 11− families showing a very similar feature as that in YBCO. It was generally understood as a crossover from an elastic to plastic vortex motion regimes.\(^1\)\(^4\)\(^1\)\(^0\)\(^1\)\(^1\)\(^2\)\(^3\)\(^2\)\(^0\). Additionally, unconventional Meissner effect was observed in \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) and \( \text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2 \) indicating a novel field enhanced pairing strength in the iron pnictide superconductors.\(^2\)\(^1\). In this Letter, we report the magnetization and relaxation in optimally doped \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \). Beside the second magnetization peak, for the first time we observed a third magnetization peak in high temperature region. The third peak becomes more pronounced when the relaxation rate is low. The occurrence of three magnetization peaks is attributed to the co-existence of large scale strong pinning centers and weak collective pinning centers. The second peak characterized by some novel features may be categorized as a new type of vortex phase transition.

The high quality single crystal was prepared by the self-flux method.\(^2\)\(^2\). The superconducting (SC) transition temperature is up to 39 K as determined from the magnetization measurements. The crystallization and chemical composition of our sample were checked by X-ray diffraction and energy dispersive X-ray micro-analysis, both show perfect quality. The magnetization were measured by the sensitive vibrating sample magnetometer (PPMS-based, Quantum Design) at the vibrating frequency of 40 Hz with the resolution better than \( 1 \times 10^{-6} \text{emu} \). The magnetic field sweeping rate can be varied from 0.5 \text{Oe/s} to 700 \text{Oe/s}. The magnetic field \text{H} is parallel to c-axis of single crystal during the measurements.

Fig.1 shows the magnetization-facing-loops (MHLs) in different temperature regions with the magnetic field sweeping rates of 50 Oe/s and 200 Oe/s. The symmetric MHL curves suggest that the bulk pinning instead of surface barrier dominates in the sample. At 2 K, the MHLs exhibit flux jumps (avalanche effect) near zero field which has been observed in many other superconductors.\(^1\)\(^4\)\(^2\)\(^3\)\(^4\) and was attributed to
FIG. 1: (color online) The MHLs measured at different temperatures with the magnetic field sweeping rates of 50 Oe/s and 200 Oe/s.

the magneto-thermal instabilities[24]. Above 5 K, a magnetization peak appears near the zero field in the MHLs and it evolves into a sharp structure at high temperatures. This sharp central magnetization peak may be understood as an evidence of strong pinning[16, 17]. When the magnetic field is swept back to zero, the Bean critical state model would assume a flat MHL near zero field if \( J_c(H) \) is a constant with \( J_c \) the critical current density. The sharp magnetization peak may suggest that the critical current density is getting stronger when the field is approaching zero. In moderate temperature region, after the first central peak, the magnetization increases with the magnetic field leading to the so-called second-peak (SP) effect. The SP shifts to lower fields with increasing temperature and becomes more obvious with more slow field sweeping rate. Finally, a third peak emerges in high temperature region (see the data at 32.5 K) and it exhibits the similar temperature dependence of the SP. For iron based superconductors, there has no report about three characteristic peaks in MHLs.

The MHLs at 33.5 K and 35 K are presented in Fig.2 (a) and (b). Three characteristic peaks can be observed easily in both curves. With the fast sweeping rate, the SP exhibits a step-like behavior (kinky shape), while it becomes more visible (peak shape) with slower sweeping rate. The position of the SP shifts slightly with different sweeping rates at the same temperature which causes the crossing of the MHL curves near SP (shown in Fig.2 (b)). To further investigate the vortex behavior, we calculated the corresponding dynamic magnetization-relaxation rate via

\[
Q = \frac{d \ln j_s}{d \ln (dB/dt)} = \frac{d \ln (\Delta M)}{d \ln (dB/dt)},
\]

where \( j_s \) is the transient superconducting current density, \( dB/dt \) is the field sweeping rate[24]. Fig.2(d) shows the magnetic field dependence of \( Q \) at 35 K. Near the
valley between the central peak and SP, a moderate relaxation with Q = 6% is observed. With increasing the magnetic field, a clear lowering down of the magnetic relaxation is observed near the edge of the SP. Interestingly the relaxation rate Q sometimes shows a negative value, which we think is not a true effect. It is induced by the above-mentioned crossing of the MHLs measured at different sweeping rates, indicating a very slow relaxation rate near the SP. Such low Q-value and the crossing effect of the MHLs have never been observed in other superconductors, this may suggest the very damped vortex motion and strong pinning. We thus categorize the second peak here as a new type vortex phase transition.

Further increasing the magnetic field, Q rises to a moderate value again and a plateau of Q can be observed below the third peak (TP) field in MHL. In high field (beyond the TP) region, the Q rises up drastically and the shear modulus $C_{66}$ drops down quickly. The vortex dynamics near the third peak is actually quite similar to the second-peak observed either in YBCO or Bi-2212.

In order to further investigate the vortex behavior and check the relaxation rate derived from the dynamic relaxation method, we also did the traditional magnetization-relaxation measurements on this sample. The time (t) dependence of the non-equilibrium magnetization (M) at 33.5 K are shown in Fig.3(a). It shows that, the slow time decay of non-equilibrium magnetization is observed near 2.5 T which is exactly corresponding to the SP in MHL. Based on the data of M vs. t, we get the traditional magnetization relaxation rate defined as $S = -dln(M)/dt$ in the time window of 100 s to 7200 s. Fig.3(b) shows the magnetic field dependence of both S and Q at 33.5 K. The data acquired by the two different methods agree with each other very well. The abnormal minimum corresponding to the SP can be observed both in the data of S(H) and Q(H).

Concerning the second peak effect in Bi-2212, different explanations were given. It was regarded as a consequence of phase transition between the low field Bragg glass and the high field vortex glass, which is supported by neutron diffraction, Hall probe, magneto-optical imaging technique and other measurements[27–30]. It shows that the SP in MHL has strong time dependence, implying a dynamical character (transient vortex profile) of the SP effect[31–34]. In our measurements, the similar time dependence of SP is found: more slower sweeping rate, more obvious the SP effect (shown in Fig.2 (b)). This may be understood in the similar way: large scale strong pinning centers separate the vortex system into domains, between the domains the vortex dynamic is dominated by the small scale or point-like disorders. In Fig.2(d), we separate the vortex dynamics into five regions due to the presence of the strong and point-like pinning centers. Region-I near the central peak is characterized by the strong pinning to very diluted vortices. In region-II, the vortex number is getting more than that of the large scale pinning centers, therefore the relaxation rate is moderate. When it is in the SP region, both the large scale and the point-like disorders behave as effective pinning centers and the vortices may be highly entangled, therefore the
relaxation rate is very low. A further increase of the magnetic field will make the vortex system more and more dense. It is in this region (region-IV), the vortex entity between the large scale pinning centers starts to shear, leading to a finite relaxation. But the dislocations are still quite limited and the critical current density are still growing up. Beyond the TP, large number of dislocations are proliferated and plastic motion occurs. Therefore the multiple magnetization peaks in the MHL are induced by cooperative interactions between the vortices, the large scale pinning centers and the point-like disorders.

Based on the Bean critical state model, we calculate the superconducting current density and shown in Fig.4(a), the corresponding relaxation rate Q are shown in Fig.4(b). The strong temperature dependence of the dip in Q(H) corresponding to SP can be observed. In Bi-2212 the SP shows weak temperature dependence in low field region and disappear at the melting line. This validates the conclusion that the SP in Bi-2212 in induced by the quenched disorder. But in our measurements, the unique temperature dependence of the SP is observed which suggests a different origin. In some experiments on YBCO, the so-called third magnetization peak was also reported. Later on it was discovered that this was induced by the vortex channelling along twin boundaries. This effect is thus sensitive to the angle between the orientation of twin boundaries and applied field. We have looked into this possibility by measuring the MHLs by fixing the angle between the magnetic field and c-axis constant, but with different in-plane angles respect to the field. We found the same results for all different in-plane angles. The TP accompanied by the instant upturn in Q shifts to the low field with increasing temperature, which is similar to the SP observed in YBCO, and some iron based superconductors, such as the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and LiFeAs. Below the TP, the Q keeps a relative low value which suggests the elastic motion of vortices.

Fig.5 shows the vortex phase diagram based on the Q values. The very low relaxation can be observed in a narrow region (the blue region shown in Fig.5) which is corresponding to the SP. The third-peak field H$_{TP}$ indicates the crossover from the elastic motion to plastic motion of vortices. In iron based superconductors, the disordered vortex structure in a large scale was observed in local measurements, which suggests strong pinning effect. In our measurements, the multiple magnetization peaks and the novel vortex dynamics can be interpreted as cooperative interactions between the vortices, the large scale pinning centers and weak but dense point-like disorders. The large scale pinning centers are probably induced by the domain walls between the superconducting and the fluctuating antiferromagnetic regions. The point-like disorders are given by the dopants, like the random distributions of Ba/K. It would be very interesting to check whether the proposed two different kinds of the pinning centers, namely the large scale strong pinning centers, and the point-like disorders are really present and play the important role for the vortex pinning.

In summary, for the first time, three characteristic
magnetization peaks are observed in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ superconductors. The second peak-effect is observed in the intermediate temperature region which is accompanied by diminishing vortex motion. In high temperature and high field regions, a third magnetization peak emerges and the vortex motion is getting drastically. The abnormal MHLs with three distinct magnetization peaks can be understood by the model with large scale strong pinning and weak collective pinning centers.

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