Doping dependence of the pinning efficiency in K-doped Ba122 single crystals prior to and after fast neutron irradiation

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Abstract
A sharp peak was observed in the doping dependence of the critical current density, \(J_c\), in potassium doped Ba122 single crystals. This behavior is in contrast to the doping dependence of the transition temperature, \(T_c\), which varies much more smoothly around its maximum. We performed fast neutron irradiation on the crystals in order to find out whether the \(J_c\) peak results from intrinsic properties or the particular defect landscape. Fast neutrons are known to introduce defects up to a size of a few nanometers, which have proven to be more efficient for flux pinning than the crystallographic defects in the pristine crystals. We find that the peak in \(J_c\) shifts to higher doping levels after the irradiation, broadens, and roughly follows the shape of the \(T_c\) curve. Moreover, a power law between \(J_c\) and \(T_c\) is observed in the irradiated crystals, which can be explained by relations between fundamental parameters and \(T_c\) observed previously in iron-based superconductors. This power law does not hold for the pristine crystals which indicates that the doping dependence of \(J_c\) results from an enhanced pinning efficiency in the under-doped area of the phase diagram.

Keywords: iron-based superconductors, critical current density, neutron irradiation, pinning efficiency

(Some figures may appear in colour only in the online journal)

1. Introduction
Iron-based superconductors of the BaFe\(_2\)As\(_2\) (Ba122) family have a rich and diverse phase diagram which is not entirely understood at the moment. The phase diagram exhibits an antiferromagnetic phase which is suppressed by doping with cobalt, phosphorous or potassium. A superconducting phase emerges in a doping range where it coexists with antiferromagnetism. The superconducting transition temperature \(T_c\) increases, peaks, and decreases again upon further doping forming a superconducting dome. Especially, the doping range with the coexistence of antiferromagnetism and superconductivity is of great interest. It is not clear at present how these two phases occur together, but there is evidence that they are in a microscopic coexistence [1].

A quantity which is of interest, not just for applications, but also for the understanding of the microscopic (pinning) landscape is the critical current density, \(J_c\). Its dependence on the dopant concentration, \(x\), should be, at first glance, similar to the doping dependence of \(T_c\). Rather contrarily, \(J_c\) shows a sharp peak while \(T_c\) varies smoothly as a function of the dopant concentration. This peak in \(J_c(x)\) was measured in Ba122 doped with potassium, phosphorous, and cobalt [2–5].
The discrepancy between \( T_c(x) \) and \( J_c(x) \) in the pristine crystals can be described in the following ways: (1) the magnetic penetration depth and the coherence length could vary as a function of doping in a similar way as \( J_c \) does; (2) the pinning efficiency resulting from the prevailing defect structure could be different for different doping concentrations; (3) an exotic mechanism like a quantum critical point (QCP) could be the reason. It is not clear yet which of the previous assumptions holds true for which doping system. For example, a sharp peak was found in the doping dependence of the magnetic penetration depth in the phosphorous doped system which was discussed as the sign of a QCP [6]. This peak in the magnetic penetration depth was used to explain the anomaly in \( J_c(x) \) [4]. Contrarily, in the cobalt doped Ba122 an increased superfluid density on twin boundaries is discussed to be the reason for the peak in \( J_c(x) \) [7].

We will focus in this study on Ba122 doped with potassium where the concentration for maximum \( T_c \) and maximum \( J_c \) is not the same. Our attempt to distinguish between the different proposed mechanisms is fast neutron irradiation. This technique is known to introduce isotropic defects up to a size of a few nanometers, which have proven to be more efficient for flux pinning than the crystallographic defects in the pristine crystals [8]. Therefore, we can assume the pinning efficiency after the irradiation to be similar for crystals with different potassium concentration. Thus, after the irradiation \( J_c(x) \) is determined only by the intrinsic parameters, namely magnetic penetration depth, and coherence length. This gives us the unique possibility to investigate the origin of \( J_c(x) \) of the pristine crystals by comparing it to \( J_c(x) \) of the crystals after the irradiation with fast neutrons.

2. Methods

The Ba\( _{88-4x} \)K\( _x \)Fe\( _2 \)As\( _2 \) single crystals were grown by the self flux method described elsewhere [9]. Single crystals with a potassium concentration ranging from \( x = 0.23 \) to \( x = 0.5 \) covering the under- and over-doped part of the superconducting dome were used. The typical size of the crystals is \( 1 \text{ mm} \times 0.8 \text{ mm} \) with a thickness of 80 \( \mu \text{m} \).

The superconducting transition temperature, \( T_c \), was determined by AC susceptibility measurements with an amplitude of 30 \( \mu \text{T} \) in a superconducting quantum interference device magnetometer (MPMS quantum design). \( T_c \) was evaluated using the 10% criterion of the saturated diamagnetic moment throughout this work.

The critical current density, \( J_c \), was obtained from magnetization measurements performed in a vector vibrating sample magnetometer. Measurements were done at constant temperatures with the magnetic field applied parallel to the c-axis of the sample. Thus, the critical current density in the ab-plane was measured. Measurements were performed at different temperatures with an applied field of up to 5 T. The \( J_c \) calculations are based on the Bean model with a numerical self-field correction where \( J_c \) and the average \( B \) are calculated self-consistently [10, 11]. The high critical current densities in the irradiated crystals result in high self-fields. In order to eliminate the influence of different self-fields, the data in the following section all refer to an average field of 1 T within the sample. Since the large field gradients within the samples potentially still influence the results, we did all the evaluations also for 2 T and found the same behavior as at 1 T. The chosen field hence seems to be a good compromise between staying in the low field limit and eliminating the influence of the changing self-field.

The fast neutron irradiation (\( E > 0.1 \text{ MeV} \)) was carried out in the TRIGA research reactor at the Atominstitut in Vienna. The fast neutron fluence applied to the samples was determined by measuring the \( ^{58}\text{Co} \) activity of a nickel foil irradiated together with the samples. The single crystals were irradiated for the same time period twice resulting in fast neutron fluences of \( 1.7 \times 10^{21} \text{ m}^{-2} \) and \( 3.4 \times 10^{21} \text{ m}^{-2} \). It is important to point out that the measurements for different fluences were carried out on the same set of crystals in order to make sure that measured differences stem solely from the irradiation and are not sample-to-sample variations. Furthermore, the crystals were irradiated together to ensure that they are all exposed to the same fluence.

3. Results

The results presented in this section can be divided into two parts: effects on the (1) critical current density and (2) critical temperature. We found a reduction of \( T_c \) for all our samples after the fast neutron irradiation which is shown in figure 1. The relative changes were used for the comparison of the \( T_c \) reduction to account for the different \( T_c \) in the pristine samples. Figure 1(a) shows the relative change of \( T_c \) with respect to the pristine \( T_c \) as a function of the K concentration. After the first irradiation step \( (1.7 \times 10^{21} \text{ m}^{-2}) \) the reduced reduction of \( T_c \) is almost constant for all samples. The data after irradiation to \( 3.4 \times 10^{21} \text{ m}^{-2} \) shows more scattering, but no clear trend can be observed. (However, the smallest reduction of \( T_c \) is measured around the optimal doping concentration [12].) Figure 1(b) shows all three AC susceptibility measurements for the crystal with 30% potassium concentration as an example. The width of the transition does not change after the irradiation implying that the quality of the crystals did not change. Moreover, the value of the magnetic moment in the superconducting phase is similar for all measurements which is an indication for an unchanged superconducting volume. The \( m(T) \) curves look similar for the other crystals. This confirms that we just introduced nanometer size defects and did not cause any further damage to the crystals by the irradiation procedure.

The critical current density, \( J_c \), and its doping dependence is plotted in figure 2. Figures 2(a) and (b) show \( J_c \) at \( B = 1 \text{ T} \) for the pristine and the irradiated crystals, respectively. The critical temperature is plotted in these figures for comparison. \( J_c(x) \) shows a distinctively different shape than \( T_c(x) \) in the pristine crystals as reported previously [2, 3]. This functional behavior remains the same for the different temperatures shown in figure 2(a). After the irradiation, \( J_c \) increased drastically due to the introduction of a pinning
landscape which is more efficient than the intrinsic defects of the crystals. Moreover, $J_c(x)$ changes its shape and resembles now the shape of $T_c(x)$ (see figure 2(c)). This change of shape is shown in figure 2(c) where $J_c(x)$ is plotted before and after irradiation at 10 K and 1 T. The effect of fast neutron irradiation on the doping dependence of the critical current density can be summarized in the following way: The maximum $J_c$ shifts to higher potassium concentrations after irradiation and the peak in $J_c(x)$ broadens and therefore resembles closer the shape of $T_c(x)$. It is also noticeable that this broadening and shifting of the peak already occurred after the first irradiation to a fluence of $1.7 \times 10^{21} \text{ m}^{-2}$. The second irradiation step resulted in a further increase of $J_c$ which is similar over the whole doping range.

4. Discussion

The difference between $J_c(x)$ in Ba122 before and after fast neutron irradiation is interesting but has not been investigated in details so far. The critical current properties can be separated into flux pinning effects and effects resulting from intrinsic parameters of the mixed state by expressing the critical current density as $J_c = \eta J_d$ where the pinning efficiency, $\eta$, accounts for flux pinning and the depairing current density, $J_d$, for the...
intrinsic superconducting parameters. $J_d$ is a function of the magnetic penetration depth, $\lambda$, and the coherence length, $\xi$: 

$$J_d = \frac{\eta}{2\sqrt{3}\pi n_0 \lambda^2}.$$ 

The depairing current density can be re-written as $J_d \propto \frac{B_c}{\lambda}$ where $B_c$ is the thermodynamical critical field. The magnetic penetration depth and the thermodynamic critical field are related to the critical temperature by power laws as reported in literature. The Umura-plot determines the dependence for $\lambda$ which is $\lambda \propto T_c^{0.5}$ [13]. The relation for $B_c$ was extracted from thermal capacity measurements and found to be $B_c \propto T_c^{0.75}$ [14, 15]. These two relations result in a correlation between the depairing current density and the critical temperature: $J_d \propto T_c^{2.25}$.

The pinning of flux lines in pristine crystals happens on defects in the crystal which can also result from the potassium atoms introduced into the lattice as doping. Therefore, a different pinning efficiency in the crystals having a varying potassium concentration is a priori expected. Irradiation of the samples with fast neutrons introduces a defect landscape which is more efficient for flux pinning than the defects of the pristine crystals and leads to an increase of the critical current density. This was shown for several iron-based superconductors irradiated with different particles [8]. Since the radiation induced defect structure is essential the same in all crystals a constant pinning efficiency, $\eta$, governed by the introduced defects will be assumed in the following. $J_c$ is consequently directly proportional to the depairing current density after irradiation and hence proportional to $T_c^{2.25}$. It is important to emphasize that the calculations of $J_d$ from above hold also true for the pristine crystals. Thus, they also exhibit a power law between $J_d$ and $T_c$, but not between $J_d$ and $T_c$, because the pinning efficiency in the pristine crystals likely depend on the dopant concentration.

The critical current density at a reduced temperature $t = \frac{T}{T_c} = 0.3$, which corresponds to approximately 10 K for the optimally doped crystals, and 1 T of the pristine crystals and the crystals after each irradiation step is plotted in figure 3(a) as a function of the critical temperature. The reduced temperature is used to ensure a proper comparison of the crystals despite their different $T_c$. However, the same analysis done at a fixed temperature of 10 K led to very similar results. The difference between the pristine and the irradiated crystals can be easily seen in this double logarithmic representation. The pristine crystals show no correlation between $J_c$ and $T_c$ which is a result of the difference in shape between $J_c(x)$ and $T_c(x)$. The correlation between $J_c$ and $T_c$ does not hold because the as-grown pinning centers are responsible for the critical current density in the pristine crystals and the pinning efficiency is not identical in all samples. Contrarily, the irradiated crystals show a linear relation in the double logarithmic plot, thus the correlation is a power law. A closer look at figure 3(a) shows that the correlation already holds after irradiation to a fluence of $1.7 \times 10^{21}$ m$^{-2}$. Further irradiation to a fluence of $3.4 \times 10^{21}$ m$^{-2}$ results in a similar increase of the critical current density with respect to the previous irradiation for all crystals. This shows that irradiating with $1.7 \times 10^{21}$ m$^{-2}$ introduces already a pinning landscape that dominates the flux pinning properties.

The correlation between $J_c$ and $T_c$ of the irradiated crystals can be described by the predicted power law $J_c \propto T_c^{2.25}$ depicted by the red line in figure 3(b). This red line results from a fit of $J_c = a \cdot T_c^{2.25}$ to the irradiated potassium doped samples with $a$ as the only fit parameter. The correlation applies for the irradiated crystals as the dominant pinning centers are the defects introduced by the fast neutron irradiation resulting in a constant pinning efficiency.

This finding raises the question if this correlation is restricted to potassium doped Ba122. Therefore, available data on irradiated optimally cobalt, potassium and phosphorous doped Ba122 crystals were added to figure 3(b) [16]. All the data points follow the predicted power law behavior of $J_c(T_c)$ shown by the red line. Our results hence indicate that the derived relation is independent of the dopant in Ba122.

Moreover, it is possible to calculate the constant pinning efficiency of the irradiated crystals underlying the red line in figure 3(b). The calculation was done for the crystal with 40%
potassium concentration as the required parameters are reported in literature. The result holds also for all other crystals because of the constant pinning efficiency. First, the depairing current density of the crystal was calculated using the superconducting coherence length, $\xi = 1.4$ nm [2], and the magnetic penetration depth, $\lambda = 230$ nm [17]. This results in a depairing current density of $13.6 \times 10^{11}$ A m$^{-2}$. Next, the pinning efficiency was calculated from $\eta_{\text{eff}} = \frac{a^{2} \cdot T_{\text{c}}}{\xi}$, where $T_{\text{c}}$ and $a$ are the critical temperature of this crystal and the fit parameter obtained with the data shown in figure 3(b), respectively. This leads to a pinning efficiency of 3.2% for all the samples after fast neutron irradiation. Note that similar pinning efficiencies were found in various compounds of the cuprate superconductors after fast neutron irradiation.

The relation $J_{c} = a \cdot T_{c}^{2.25}$ enables the calculation of $J_{c}$ from the measured values of $T_{c}$. Figure 4 shows these calculated $J_{c}$ values alongside with the measured values. $J_{c}(x)$ of the irradiated samples, shown in figure 4(a), follows closely the calculated $J_{c}(x)$ curve except for the sample with a potassium concentration of 33%. This small peak is preserved at all other measured temperatures as shown in figure 2(b). It needs to be clarified, if this point can be considered as a normal sample-to-sample variation or an indication of physics beyond our model.

Contrarily, the correlation between $J_{c}$ and $T_{c}$ does not hold for the pristine crystals (see figure 4(b)). This is a consequence of a pinning density using the relation we found for the irradiated crystals and their constant pinning efficiency $\eta_{\text{eff}}$: $J_{d} = \frac{a}{T_{c}} \cdot T_{c}^{2.25}$. The result $\eta_{\text{pris}} = \frac{J_{d}}{J_{c}}$ is shown in figure 4(c).

The pinning efficiency is highest for the under-doped crystals and starts to decrease upon further doping. The enhancement of the pinning efficiency takes place in the same doping range where antiferromagnetism and the orthorhombic phase are present. It was discussed previously that the twin boundaries in the orthorhombic phase act as pinning centers [5]. This would explain this enhancement, but a previous study on cobalt doped Ba122 showed that the peak in $J_{c}(x)$ does not disappear after detwinning the samples by applying uniaxial pressure [18]. This peak can be directly associated with an enhancement of the pinning efficiency. Thus, if the twin boundaries were the reason for the enhanced pinning efficiency in the under-doped region the peak in $J_{c}(x)$ should disappear after detwinning.

Another candidate for enhancement of the pinning efficiency could be the antiferromagnetism which is present in the same doping range. Evidence has been found for microscopic coexistence of magnetism and superconductivity in potassium doped Ba122 using muon spin rotation and infrared spectroscopy [1]. These measurements contradict possible microscopic magnetic inclusions in the superconductor which could act as pinning centers. However, defects in the antiferromagnetic lattice which lead to a non vanishing magnetic moment in a certain region could contribute to pinning. Flux lines could then be pinned in these regions by electromagnetic interaction. Moreover, magnetic domain walls could result in non vanishing magnetic moments in certain regions and thus act as pinning centers.

All the attempted explanations above assume that the peak in $J_{c}(x)$ results from pinning and not from superconducting parameters like the magnetic penetration depth or the coherence length varying for the different dopant concentration in the crystals. If the peak in the pristine $J_{c}(x)$ does not result from pinning but from a peak in these superconducting parameters, it should still be visible after the irradiation since the additionally introduced pinning centers would increase $J_{c}$ but the shape of $J_{c}(x)$ would still be governed by the (unchanged) superconducting parameters, which is in contradiction to our experimental findings.

5. Conclusion

We have carried out measurements of the critical temperature and the critical current density on a set of potassium doped Ba122 single crystals with the doping level ranging from $x = 0.23$ to $x = 0.50$ prior to and after fast neutron irradiation. A correlation between $J_{c}$ and $T_{c}$ was found after irradiation to a fast neutron fluence of $3.4 \times 10^{11}$ m$^{-2}$ which did not prevail in the pristine crystals. This correlation also holds for literature data of irradiated Ba122 doped with phosphorous and cobalt. Thus,
this behavior seems to be universal for Ba122. The measured correlation between $J_c$ and $T_c$ in the irradiated samples is explained by a constant pinning efficiency and power law between $J_d$ and $T_c$. This power law holds true for the pristine and irradiated crystals but can be assessed only in the latter case because the pinning efficiency is constant there for all dopant concentrations. Furthermore, we calculated the pinning efficiency of the pristine crystals as a function of doping, which showed a sharp maximum in the under-doped region where antiferromagnetism and superconductivity coexist. A further investigation of the pinning mechanism in the doping range where the pinning efficiency peaks could reveal methods to increase the performance of technological superconductors and help to understand the underlying physics responsible for pinning in these materials.

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