

Neutron irradiation of $\text{SmFeAsO}_{1-x}\text{F}_x$

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Abstract

$\text{SmFeAsO}_{1-x}\text{F}_x$ was irradiated in a fission reactor by a fast ($E > 0.1$ MeV) neutron fluence of $4 \times 10^{21} \text{ m}^{-2}$. The introduced defects increased the normal state resistivity due to a reduction in the mean free path of the charge carriers. This leads to an enhancement of the upper critical field at low temperatures. The critical current density within the grains, J_c , increases upon irradiation. The second maximum in the field dependence of J_c disappears and the critical current density becomes a monotonically decreasing function of the applied magnetic field.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The discovery of superconductivity in the iron pnictides [1] is interesting from a theoretical point of view, but this new class of superconductors could also become important for applications. The fundamental superconducting parameters are promising. The transition temperature reaches about 55 K [2–4], which is not as high as in most cuprates but is significantly higher than in the technologically relevant superconductors NbTi and Nb₃Sn or in MgB₂. The upper critical field, B_{c2} , is extremely large (>50 T) [5–10] and thermal fluctuations seem to be less important than in the cuprates [7, 11], where loss free currents are restricted to fields far below the upper critical field, at least at elevated temperatures. Zero resistivity was demonstrated at fields close to $B_{c2}(T)$ in pnictide single crystals [8].

Irradiation techniques are a powerful tool for assessing the influence of defects on superconductors, because they allow one to investigate the same sample prior to and after the irradiation, which excludes problems of sample to sample variations. In particular, neutron irradiation was used in extended studies of the influence of disorder in MgB₂ [12–15], including the demonstration of the disappearance of two band superconductivity due to interband scattering at high levels of disorder [16]. An increase in the upper critical field and a reduction of its anisotropy were reported and are expected theoretically. A similar behaviour of B_{c2} was also found in the A15 compounds [17] and a reduction of anisotropy was reported for the cuprates (Hg-1201) [18].

The neutron-induced defects are also highly suitable for investigating flux pinning. This was done successfully in V₃Si [19], the cuprates [20, 21] and MgB₂ [22].

The present contribution reports on a first neutron irradiation experiment with the new FeAs based superconductors. Changes of the reversible and irreversible superconducting properties were found to be similar to other superconducting materials.

2. Experimental details

The $\text{SmFeAsO}_{1-x}\text{F}_x$ sample was prepared at the National High Magnetic Field Laboratory, Florida State University. The starting materials of As, Sm, Fe, Fe₂O₃ and SmF₃ were mixed and pressed into a pellet, wrapped with Nb foil and sealed in a stainless steel tube. The sealed sample was heat treated at 1160 °C for 6 h in a high temperature isostatic press under a pressure of 280 MPa. The main phase of the sample is $\text{SmFeAsO}_{1-x}\text{F}_x$, with a grain size of 10–15 μm. The impurity phases include SmAs, SmOF and FeAs. The size of the sample used for this study was $1.1 \times 1.7 \times 3.7 \text{ mm}^3$.

Neutron irradiation was performed in the central irradiation facility of the TRIGA-Mark-II reactor at the Atomic Institute in Vienna. The sample was sealed into a quartz tube and exposed to the neutron flux for 14 h and 38 min, corresponding to a fast ($E > 0.1$ MeV), thermal ($E < 0.55$ eV) and total neutron fluence of $4 \times 10^{21} \text{ m}^{-2}$, $3.2 \times 10^{21} \text{ m}^{-2}$ and $1.1 \times 10^{22} \text{ m}^{-2}$, respectively [23]. Neutrons transfer their energy to the lattice atoms by direct collisions. The transferred energy must exceed the binding energy of

the lattice atom to displace it, thus only fast neutrons lead to defects. No indirect defect-producing mechanism (e.g. an induced α -emission in MgB_2 [14, 24] or a fission reaction in uranium doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [25]) exists.

The smallest resulting defects are single displaced atoms (point defects), but larger defects might also occur. In high temperature superconductors, the largest defects are so-called collision cascades [26]. These spherical defects are amorphous with a diameter of 2–3 nm; the surrounding strain field enlarges the defect to about 5 nm. Similar defects were also found in MgB_2 [22, 27]. The actual size, morphology and density of the defects in $\text{SmFeAsO}_{1-x}\text{F}_x$ are currently unknown. However, the defects should be randomly generated, leading to a homogeneous defect density on a macroscopic length scale. Self-shielding effects can be neglected, since the penetration depth of fast neutrons is estimated to be a few centimetres, which is much larger than the sample dimensions. Only neutrons of low or intermediate energies are shielded efficiently because of the large neutron cross section of samarium at these energies, but the corresponding reactions or collisions are not expected to produce any defects.

The resistivity was measured at various fixed fields while cooling at a rate of 10 K h^{-1} with an applied current of 10 mA. Current and voltage contacts were made by silver paste. The distance between the two voltage contacts was about 1 mm.

The transition temperature at each field, $T_c(B)$, was defined as the temperature where the resistivity drops to $0.95\rho_n(T)$ ($\rho_n(T)$ was extrapolated linearly from its behaviour between 55 and 60 K). The upper critical field, $B_{c2}(T)$, was obtained by inversion of $T_c(B)$. The transition width, ΔT , is defined by the difference between T_c and the temperature where $\rho(T)$ becomes $0.05\rho_n(T)$.

Magnetization loops at various temperatures were recorded in a commercial 7 T SQUID magnetometer. The field was always oriented parallel to the smallest sample dimension (transverse geometry). The ac susceptibility at 33 Hz was measured with an amplitude of $30 \mu\text{T}$ at various fields and temperatures in order to estimate the shielding fraction. The demagnetization factor was calculated numerically for the actual sample geometry.

3. Results and discussion

The transition temperature decreases after irradiation, from 53.6 to 53.2 K, which is similar to the decrease found in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [28] but less than in the thallium based cuprate superconductors [29] at the same fluence. This decrease is ascribed to d-wave superconductivity in the cuprates [30, 31], the reason for the reduction in T_c of $\text{SmFeAsO}_{1-x}\text{F}_x$ is currently unknown. A moderate amount of non-magnetic impurities does not reduce T_c in isotropic single band superconductors, thus anisotropy [32–34] or multiband superconductivity [35–38] are possible candidates for explaining this decrease. Recently, a suppression of T_c by impurity scattering was also predicted for extended s-wave superconductors [39].

The transition width is rather large ($\Delta T = 3.8 \text{ K}$ at 0 T) and significantly broadens in magnetic fields, as can be seen

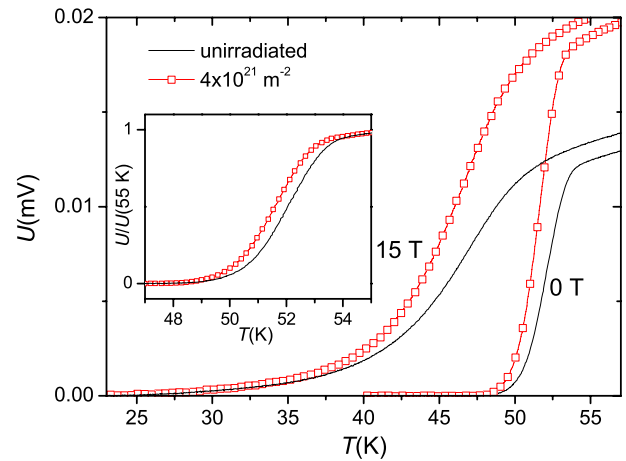


Figure 1. Resistive transitions prior to and after neutron irradiation. The inset shows the zero field transition normalized to the voltage at 55 K. The transition broadens with magnetic field, but no broadening results from the irradiation. The resistance is significantly enhanced by irradiation.

in figure 1. It remains unchanged after irradiation, which confirms the homogeneous distribution of the introduced defects.

The measured resistivity increases due to the introduction of the defects, from about 235 to $355 \mu\Omega \text{ cm}$ ($\sim 50\%$) at 55 K. We also observe an increase of the phonon contribution to the resistivity between 50 and 300 K, $\Delta\rho := \rho_n(300 \text{ K}) - \rho_n(55 \text{ K})$, from 470 to $650 \mu\Omega \text{ cm}$, which is expected to remain unchanged in single band conductors but can be altered in multiband conductors [40] (e.g. $\text{SmFeAsO}_{1-x}\text{F}_x$). Alternatively, the increase in $\Delta\rho$ could result from a reduced connectivity [41] or simply from changes in the distance between the voltage contacts, which had to be removed for the irradiation and renewed afterwards. A simple correction of these effects (keeping $\Delta\rho$ constant) reduces the increase in resistivity due to the irradiation to about 10%. The experimental accuracy (distance between voltage contacts) does not allow a final conclusion about the absolute change of the resistivity or which scenario applies. A reliable indication of the change in resistivity is given by $\rho_n(300 \text{ K})/\rho_n(55 \text{ K})$, which is independent of possible geometrical errors and which decreases from 3.03 to 2.83. Thus, we conclude that the residual resistivity is enhanced. Note that the resistivity is strongly temperature dependent above T_c . Therefore, the residual resistivity ρ_0 is smaller than $\rho_n(55 \text{ K})$ but cannot be assessed. The residual resistivity ratio ($\text{RRR} = \rho_n(300 \text{ K})/\rho_0$) is certainly larger and might be changed much more significantly due to the irradiation. This is also true for the relative change in residual resistivity. We emphasize that the resistivity and the RRR might be influenced by the impurity phases.

A decrease in the mean free path of the charge carriers is expected to result in an increase of the upper critical field. Indeed, the slope $-dB_{c2}/dT$ increases after irradiation (figure 2), but this cannot compensate for the small reduction ($\sim 0.4 \text{ K}$) of the transition temperature in the investigated field range ($\leq 15 \text{ T}$). However, the enhanced slope indicates that the

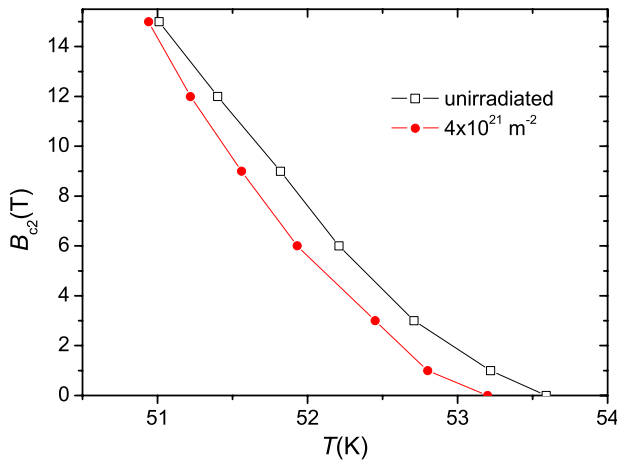


Figure 2. Change of the upper critical field after neutron irradiation.

upper critical field will be enhanced at low temperatures. The slope is nearly constant above 6 T ($-dB_{c2}/dT = 7.5 \text{ T K}^{-1}$) before the irradiation and a positive curvature can be observed up to 15 T after the irradiation, with a maximum slope of 10.7 T K^{-1} between 12 and 15 T. This corresponds to an enhancement by about 40%. Note that this enhancement is incompatible with d-wave superconductivity, for which impurity scattering is predicted to *decrease* $-dB_{c2}/dT$ [42].

The hysteresis loop at 5 K prior to and after neutron irradiation is plotted in the upper panel of figure 3. The paramagnetic background, which arises most probably from impurity phases, slightly decreases, but the hysteresis increases. The latter indicates an increase in the critical current densities, which are estimated from the Bean model in the lower panel. Zero intergranular currents (see below) were assumed and cuboidal grains with typical dimensions of $10 \mu\text{m}$, as found by backscattered electron imaging. The solid and open symbols refer to the unirradiated and irradiated state, respectively. The critical current density, J_c , increases more significantly at higher temperatures and the ‘fishtail’ effect [5, 43] disappears after neutron irradiation, i.e. J_c decreases monotonically with field. This is strikingly similar to the changes found in single crystalline [20, 44] and melt textured [45, 46] cuprate superconductors and indicates that pinning in the unirradiated sample is between weak (ordered flux line lattice) and strong (disordered flux line lattice) pinning. Thus, an order–disorder transition takes place at intermediate fields which leads to the second peak (or fishtail effect) in the magnetization curve [22, 47–50]. The neutron-induced defects are strong enough to deform the flux line lattice plastically over the whole field range. However, the defects resulting from neutron irradiation are usually not the most efficient pinning centres. For instance, the critical current densities in neutron-irradiated cuprates are approximately one order of magnitude smaller than the highest reported values (in films [51], or in single crystals containing columnar defects [52]). Thus, the critical currents should be more efficiently improved in $\text{SmFeAsO}_{1-x}\text{F}_x$ by the addition of stronger pinning centres.

The interpretation of the irreversible magnetic moment, m_{irr} , in terms of J_c is quite delicate, since it is a

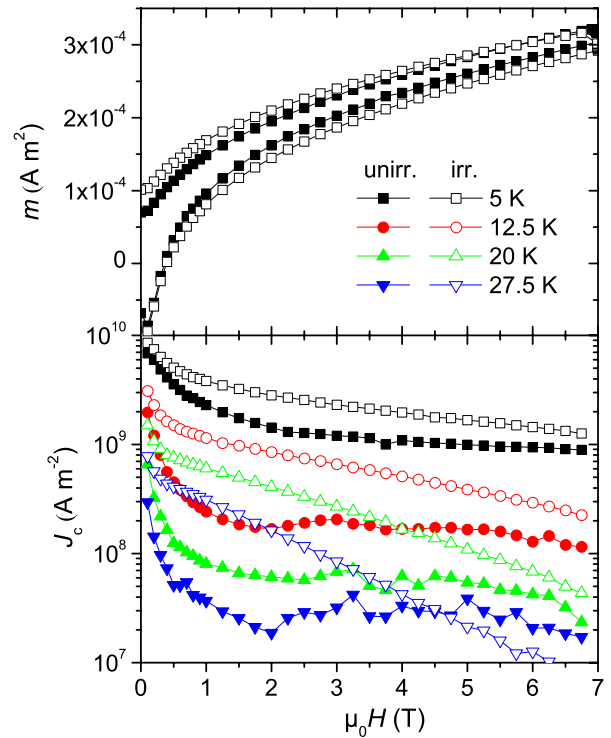


Figure 3. The width of the hysteresis loop increases after irradiation (upper panel) and the ‘fishtail’ effect disappears (lower panel).

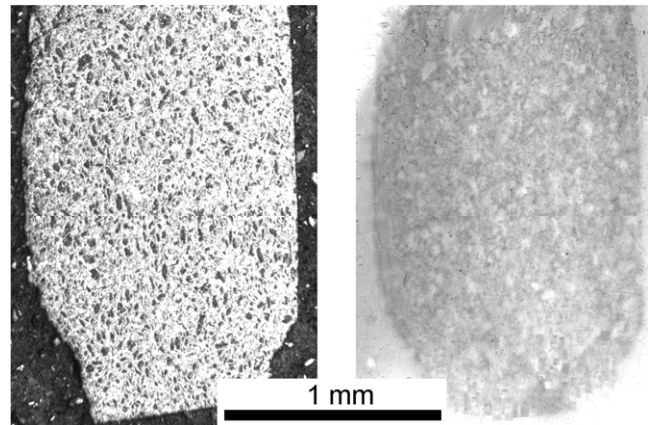


Figure 4. Optical (left) and magneto-optical (right) image of a reference sample at 6 K and 60 mT after zero field cooling. No global Bean profile is established, showing that the intergranular currents are negligible.

priori unknown, whether m_{irr} is predominantly generated by (intergranular) currents flowing around the whole sample or by (intragranular) currents shielding individual grains only. We observed a pronounced magnetic granularity by magneto-optical imaging, with no global Bean profile (figure 4, right). Thus, it is safe to assume that the irreversible magnetic moment is made up almost entirely by intragranular currents. This behaviour is in marked contrast to that seen in a high pressure treated Sm-1111 sample where clear evidence was seen of the global current flow [43, 53].

This is also demonstrated by ac susceptibility measurements. Diamagnetic shielding was nearly perfect in the present

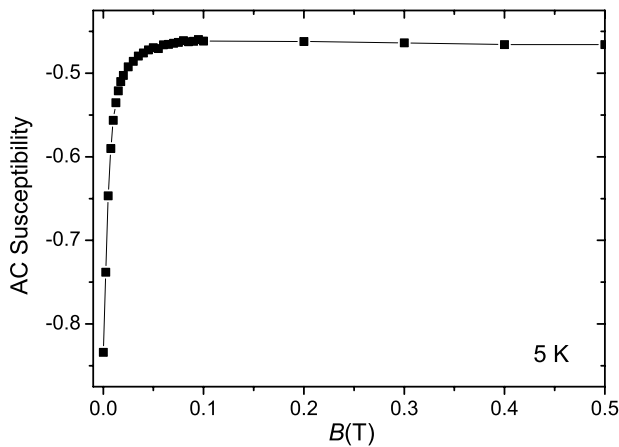


Figure 5. The ac susceptibility decreases rapidly with increasing magnetic field, since the grains decouple.

sample at zero magnetic field. The data in figure 5 refer to 5 K. The susceptibility was calculated by assuming a demagnetization factor of 0.58. The slightly smaller value at zero applied dc field (-0.83 instead of -1 in the ideal case) can result either from a residual magnetic field or from geometrical imperfections. The ac susceptibility decreases by a factor of nearly 2 by applying a dc field of only 60 mT and remains constant at higher fields. The rapid decrease at low fields reflects the decoupling of the individual grains and the small ac field penetrates the whole sample along the grain boundaries at fields above about 60 mT, when the intergranular currents become negligible.

4. Conclusions

The introduction of disorder by fast neutron irradiation enhances the normal state resistivity and the upper critical field at low temperatures. Efficient flux pinning centres are introduced, which enhance J_c . Like in the cuprates, the second maximum ('fishtail') in the field dependence of the critical current disappears, implying that the irradiation-induced defects distort the flux line lattice plastically at all magnetic fields.

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