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The effect of columnar defects on the pinning properties of $NdFeAsO_{0.85}$ conglomerate particles

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Abstract

An oxypnictide superconductor $NdFeAsO_{0.85}$ sample with transition temperature of 46 K was irradiated with 2 GeV Ta ions at a fluence of 5×10^{10} ions cm^{-2}. High resolution transmission electron microscopy study revealed that the irradiation produced columnar-like defects. The effect of these defects on the irreversible magnetization in polycrystalline randomly oriented fragments was studied as a function of field angle and field sweep rate. We find that the critical current density is moderately enhanced at fields below the matching field (~1 T). The pinning enhancement is anisotropic and maximum along the defect direction at 35 K, but becomes more isotropic as the temperature is decreased. The creep rate is suppressed at 35 K and at fields below the matching field, indicating that the columnar defects are efficient pinning sites under these H and T conditions.

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

1. Introduction

It is hoped that the newly-discovered, iron-based oxypnictide superconductors will show promise for technological applications, where the basic requirements are to carry high currents in high magnetic fields, and to work at relatively high temperatures where the associated cooling costs are lower. The superconducting transition temperature, T_c, in these materials has been recorded up to ~55 K [1] which puts the T_c of the pnictides above that of MgB_2 superconductors ($T_c = 39$ K), but below the high temperature superconductors (HTSC) such as optimally doped YBCO ($T_c = 92$ K) and BSCCO-2223 ($T_c = 110$ K). The $NdFeAsO_{0.85}$ sample studied here is a member of the '1111' phase of oxypnictide superconductors. The 1111 phase has been shown to have superconducting critical current densities, J_c, of 10^5–10^6 A cm^{-2} (at 5 K) in single crystals [2, 3], while in a Co-doped $BaFe_2As_2$ single crystal, a member of the '122' family [4], a J_c of 10^6 A cm^{-2} (at 2 K) has also been reported. In polycrystals of 1111 the J_c is only up to 10^4–10^5 A cm^{-2} (at 5 K) [5–7], while initial attempts to make wires of 1111 phase using the powder-in-tube method [8] have also shown low intergrain $J_c \sim 10^5$ A cm^{-2} (at 5 K). So intergrain connectivity might be even more of a concern in these materials.

One way to improve J_c is by deliberately introducing defects to the crystal lattice which can then act as strong pinning sites for vortices, increasing the irreversible magnetization and suppressing magnetic relaxation. Additionally the defects can reduce the coherence length thereby increasing the field at which the irreversible magnetization disappears (H_{irr}) on warming, and so increasing $J_c(B)$.

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There have been various studies that have aimed to increase the pinning in both 1111 and 122 pnictide superconductors. The effects of neutron irradiation, which introduces point defects to a sample, have been studied for polycrystalline samples of the 1111 phase [5, 9]. It was shown that J_c is increased and the low temperature H_{c2} was enhanced in polycrystals of $SmFeAsO_{1-x}F_x$ [5]. However, the disorder induced by the neutrons also depressed T_c [9]. Very recently, the effect of heavy ion irradiation (using 200 MeV Au ions) has been investigated in single crystal 122 with composition $Ba(Fe_{0.93}Co_{0.07})_2As_2$, and has demonstrated that columnar defects were introduced and act as effective pinning centres, increasing $J_c(B)$ by almost a factor 10 compared to the unirradiated crystal [4]. From measurements of magnetic relaxation, Nakajima *et al* infer that the effective pinning energy is enhanced by a factor 2 with irradiation, which is consistent with the increased J_c and reduced vortex creep rates. The superconducting anisotropy is thought to be considerably different for 1111 and 122 compounds, with 1111 reported to be more anisotropic ($\gamma_{Hc2} \sim 5.5$ close to T_c and temperature dependent), the precise value and its temperature dependence still being discussed in the literature [10, 11]. The 122 oxypnictide systems on the other hand are less anisotropic ($\gamma_{Hc2} \sim 2$ and only slightly temperature dependent), and therefore directional pinning from correlated defects would not be expected to show any strong temperature dependence in this case [2].

Here we study the influence of columnar defects on the more anisotropic 1111 system, where columnar-like pinning defects might be thought to play a more significant role. We present data on heavy ion irradiation of $\sim$100 μm sized polycrystalline conglomerate particles of $NdFeAsO_{0.85}$. We irradiated these samples with 2 GeV Ta ions, and studied the field dependence of the magnetization and magnetic relaxation while investigating the effect of changing the field angle away from the defect direction. In a previous study we showed that unirradiated conglomerate particles of $NdFeAsO_{0.85}$ displayed imperfect intergranular connectivity with $J_c \sim 5 \times 10^4$ A cm^{-2} at 15 K [6]. We find a significant increase in J_c with irradiation, and also a temperature dependent directionality to the pinning enhancement. We discuss the pinning by columnar defects in $NdFeAsO_{0.85}$ from the view-point of pinning in the high temperature anisotropic superconductors.

2. Experimental methods

$NdFeAsO_{0.85}$ was made by high pressure synthesis as described elsewhere [1]. The material was prepared as a pressed bar and subsequently broken into platelet-like fragments with a range of sizes. We selected those fragments with an average diameter of $\sim$100 μm and thickness $\sim$50 μm. Structural characterization has shown that the particles are polycrystalline (the grain size is of the order of microns) with secondary phases of Fe_xO_y and NdO [6]. The superconducting phase is compositionally homogeneous, but the secondary oxide phases present are probably responsible for the paramagnetic background seen in magnetization curves.

A large collection of the fragments was laid flat on an aluminium sheet and stuck down with a thin coating of GE varnish. Irradiation was carried out at the GSI Centre for Heavy Ion Research in Darmstadt using a 2 GeV beam energy of Ta ions at a fluence 5×10^{10} ions cm^{-2}. The beam was directed perpendicular to the aluminium plate such that the Ta ions would be expected to penetrate the full fragment thickness. The collection of fragments was measured while still on the aluminium plate to maintain the defect orientation. The magnetization versus applied field $M(H)$ measurements were taken at sweep rate 0.5 T min^{-1} in an Oxford Instruments variable-temperature transverse vibrating sample magnetometer (TVSM) which has a rotation stage to allow the sample to be tilted with respect to the applied field direction. The dynamic creep rate, S, was obtained using the method known as 'sweep creep' [12] as first proposed by Půst *et al* [13]. The flux creep was determined at each field by taking full $M(H)$ loops at several field sweep rates between 0.1 and 0.7 T min^{-1} and using the relation $S = \mathrm{d}(\ln \Delta M)/\mathrm{d}(\ln \mathrm{d}H/\mathrm{d}t)$. The T_c in the irradiated fragments was determined from magnetization data as being $\sim$46 K, which we note is the same as T_c in the unirradiated fragments as also extracted from magnetization measurements.

3. Results and discussion

A typical $NdFeAsO_{0.85}$ fragment is shown in the optical micrograph in figure 1(a). Inspection with field-emission scanning electron microscopy (FE-SEM) in part (b) of the same figure shows that each fragment is made up of plate-like particles of homogeneous superconducting material. After irradiating a collection of these fragments with 2 GeV Ta ions we performed high resolution transmission electron microscopy (TEM) analysis to identify the effect of the irradiation. The top surface is the one that was irradiated and we then prepared cross-sections that were oriented in a plane perpendicular to the surface using a focused ion-beam (FIB). Figures 1(c) and (d) show TEM cross-sections taken 1 μm and 5 μm below the surface, respectively. Both images show the presence of collinear tracks which, after considering the thickness of the cross-sections, agree in number with the Ta ion fluence of 5×10^{10} ions cm^{-2}. By inspection, the tracks are approximately 5–10 nm wide and therefore might be expected to suitably pin vortex lines where the vortex radius is set by the superconducting coherence length measured as $\xi \sim 3.5$ nm in the closely related system $LaFeAsO_{0.89}F_{0.11}$ [14].

In figure 2(a) we show the four-quadrant M–H envelope loop at 10 K for the irradiated fragments (open circles) and a separate collection of unirradiated fragments that were taken from the bar sample (solid curve). The field was applied parallel to the irradiation direction (we define this orientation as $\theta = 0°$). The unirradiated set was additionally measured at different applied field angles and we observed no significant angular dependence. Both curves show the presence of a similar paramagnetic background which was attributed to a small amount of Nd-oxide present in the samples [6]. The curve for the unirradiated set in figure 2(a) is asymmetric about the field axis, which we associate with the pinning

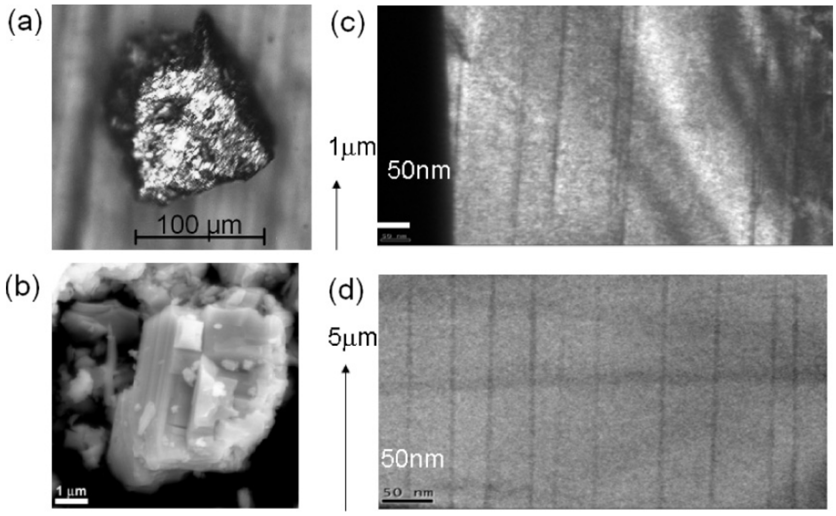

Figure 1. (a) Optical image of a typical $NdFeAsO_{0.85}$ conglomerate particle, first taken from the sintered body and then irradiated by 2 GeV Ta ions with a fluence 5×10^{10} ions cm^{-2}. (b) FE-SEM image of platelet-like $NdFeAsO_{0.85}$ particle. TEM images show (c) a cross-section taken $\sim$1 μm below the irradiated surface and (d) a cross-section taken more than 5 μm below the surface. Both (c) and (d) images show the presence of collinear columnar tracks, which are parallel to the irradiation direction (from top to bottom).

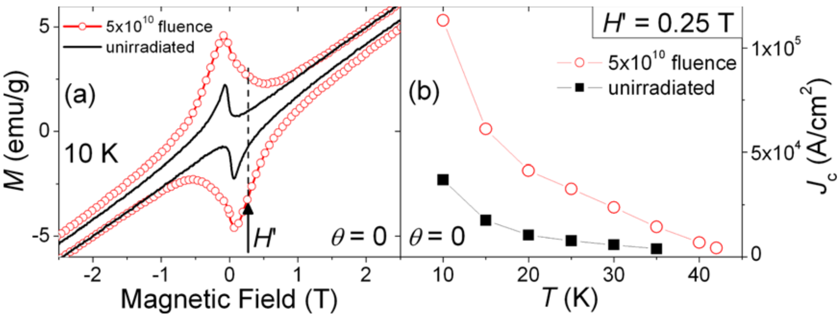

Figure 2. (a) Magnetization versus magnetic field $M(H)$ loops at 10 K on the unirradiated (black curve) and 5×10^{10} ions cm^{-2} irradiated fragments (open circles) taken at the field sweep rate 0.5 T min^{-1}; the sloping background arises from contamination by a paramagnetic second phase. The field is applied parallel to the irradiation direction—defined as $\theta = 0°$. Pinning enhancement is most notable at fields below 1 T corresponding to the matching field. (b) The critical current density at $H = H' = 0.25$ T versus temperature for the irradiated (open circles) and unirradiated sets (closed squares) for field alignment with the defect direction.

being predominantly due to surface barriers. In particular we observed slight enhancement in the sweep-rate dependence of $M(H)$ on the increasing field-leg compared to the decreasing field-leg for $H < 0.2$ T and over the limited range of sweep rates 0.1–0.7 T min^{-1}, consistent with the expected effect of surface barriers [15]. We shall present data on the dynamic creep rate later. The curve for the irradiated set in figure 2(a) showed greatest enhancement of the irreversible magnetization ΔM below the matching field of 1 T, which is when the density of vortices equals the density of columnar tracks. Note there is still a small enhancement in ΔM at fields above the matching field [16]. We extract J_c from ΔM using the Bean model and assuming that the grains within the fragments are well-connected, and that the fragments have a cylindrical geometry with average diameter 100 μm. The sample self-field is quite large, e.g. at 10 K (figure 2(a)) it reaches $\sim$0.15 T, and so introduces significant asymmetry in the $M(H)$ loops close to zero applied field; we therefore use the J_c measured at $H' = 0.25$ T to compare the irradiated and unirradiated data sets (H' is indicated by the vertical dashed line in figure 2(a)). Figure 2(b) shows the temperature dependence of J_c and shows that irradiation has enhanced J_c by a factor of 3 or 4.

In figure 3(a) we show $M(H)$ loops at 10 K on the irradiated fragments as the field is tilted from parallel ($\theta = 0°$) to perpendicular ($\theta = 90°$) to the columnar defects. The different M–H loops show a clear systematic change in magnetization with θ for fields below the matching field. At fields close to zero the curves for different angles all intersect—i.e. the remnant magnetizations are similar. This effect has been attributed to ‘flux flop’ in the HTSC [17, 18] where isotropic pinning was observed at low applied fields. The variation in ΔM with θ is greatest close to $H' = 0.25$ T, and in figures 3(b) and (c) we show this variation in $\Delta M(\theta)$ at 10 K and 35 K, respectively. As expected the ΔM is maximum for the parallel field alignment. The black squares in figures 3(b) and (c) show $\Delta M(\theta)$ for the unirradiated fragments, which confirm that there is no angular dependence in the unirradiated sample. Comparison of the $\Delta M(\theta)$ curves at 10 and 35 K shows that at 10 K there is still a significant increase in pinning with irradiation for the field perpendicular, $H_{\perp}$. At 35 K

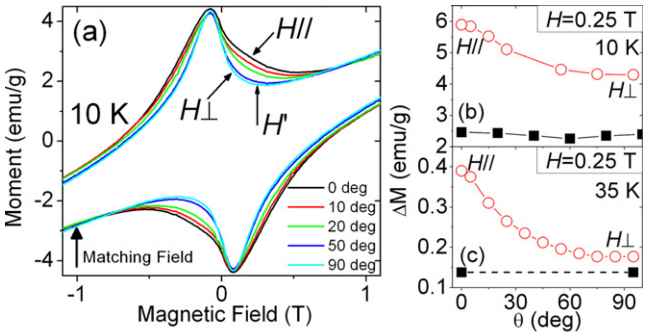

Figure 3. (a) $M(H)$ loops of the 5×10^{10} fluence irradiated fragments as the applied field is tilted from parallel ($\theta = 0°$) to perpendicular (90°) to the irradiation direction at 10 K. ΔM is extracted at the field H' indicated in (a) and plotted versus θ in (b) at 10 K and (c) at 35 K for 5×10^{10} irradiated fragments (open circles) and the unirradiated fragments (closed squares).

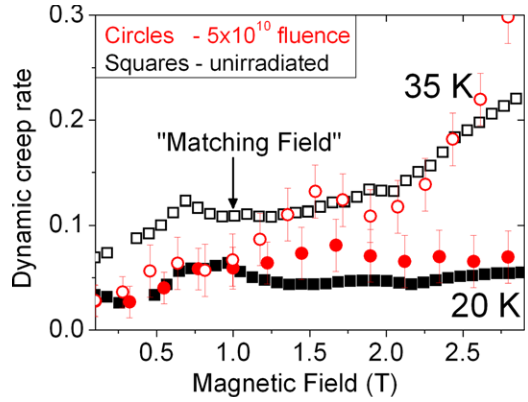

Figure 4. Dynamic creep rate, S, versus applied magnetic field at 20 K (closed symbols) and 35 K (open symbols) taken on the unirradiated bar (squares) and the 5×10^{10} irradiated fragments (circles). At 20 K the creep rate for the two samples remains similar, however at 35 K the creep in the irradiated fragments below the matching field is strongly suppressed compared to the creep in the unirradiated bar sample. The peak feature in S below 1 T for the unirradiated bar was associated with the peak effect in the $M(H)$ loop [6].

the pinning for the $H_{\perp}$ case shows little enhancement with irradiation.

It is important to compare the effect that columnar defects have on pinning in the 1111 and 122 systems with similar studies that were originally done on the HTSC superconductors such as YBCO and BSCCO [19–21]. Irradiation by heavy ions is known to have a maximum effect, as the resultant columnar defects can pin the line-like vortices along their entire length. This leads to a directional enhancement of J_c which is dependent on the angle between the applied field and the defect orientation. In particular a lock-in effect is expected for small field angles away from the defect direction and at high temperatures [22], similar to the lock-in effect of pinning by twin planes in YBCO [23]. This directional enhancement was observed down to ~20 K in YBCO crystals which have a moderate superconducting anisotropy γ, but as γ increases, (such as for highly anisotropic BSCCO crystals) there evolves a temperature crossover to a regime where the pinning enhancement is isotropic [17]. This effect was attributed either to the increasing dominance at low temperature of pinning by random (therefore isotropic) point defects, or to a 3D to 2D crossover in the vortex structure with decreasing temperature. For BSCCO the pancake vortices interact to form a line-like structure at high temperature but decouple at low temperature, and therefore lose any preference for directional pinning.

Measurements of J_c and magnetic relaxation as a function of temperature and angle have in the past [24, 25] provided insight into the type of pinning mechanism that is responsible, and addressed whether the mechanism is significantly changed by the introduction of columnar defects. Figure 3 implies that the pinning enhancement with columnar defects becomes more isotropic at low temperatures, reminiscent of pinning in YBCO and BSCCO, except that no complete crossover regime is observed for the 1111 material. In this regard the 1111 oxypnictides appear to be closer in behaviour to YBCO.

Figure 4 shows the field dependence of the dynamic creep rate at the temperatures 20 and 35 K. The magnetization signal from the set of unirradiated fragments was too small to obtain reliable creep data, however the magnetization behaviour of the fragments was characteristic of that of the bulk, bar sample [6]. Therefore, the creep data was obtained on the bar sample where a more reliable signal could be taken (the large error bars for the irradiated curve shown in figure 4 reflect that the set of fragments was much smaller in mass than the unirradiated bar sample). Figure 4 reveals that the creep rate in the irradiated fragments at 35 K is suppressed at fields below the matching field compared to the unirradiated bar. This is consistent with previous studies where the columnar defects suppressed vortex creep as the defects were effective pinning sites. At 20 K the creep rates in the irradiated and unirradiated cases are similarly low, increasing slowly with field from the low-field value ~0.03, confirming that the columns are having much less direct influence on the pinning at low temperatures; above the matching field irradiation perhaps increases the creep rate, but the data are only marginally significant.

Previous studies of magnetic relaxation [3, 26] show that the relaxation rates are large in the 1111 phase and suggest a collective vortex pinning scenario. The vortex H–T phase diagram has been mapped and this shows a large region where the melting of the vortex lattice occurs for both 1111 and 122 families [10, 27]. The vortex behaviour is described as being intermediate of YBCO and BSCCO, but perhaps closer to YBCO. However, this point is still controversial as it has also been reported [28] from measurements of the flux dynamics in the 1111 system that 2D vortex behaviour, comparable to the pancake vortex structure in BSCCO 2223 superconductor, was observed.

It is of some surprise that the columnar defects act less effectively as pinning sites in the 1111 system than in the 122 phase [4]. As the 1111 phase is considered to be more anisotropic than 122, we would have expected greater enhancement due to the columnar defects. Direct comparison is complicated however because the 1111 samples we have studied are polycrystalline, so the random orientation of the irradiation with respect to the crystallographic axes could well attenuate the overall impact of the columnar defects on pinning (we believe that the intergrain connectivity in the fragments, which would affect the magnetization, is not degraded by the irradiation).

4. Conclusions

We have studied the effect of heavy ion irradiation with 2 GeV Ta ions in polycrystalline $NdFeASO_{0.85}$ and its effect on the critical current density J_c and the flux dynamics. High resolution TEM confirms the formation of continuous collinear columnar defects. The irradiation significantly increases (factor of 3–4) the J_c to $\sim 10^5$ A cm^{-2} at 10 K. By comparison, heavy ion irradiation parallel to the c-axis of a 122 $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystal [4], increased J_c from 10^5 A cm^{-2} (at 10 K) in the unirradiated material to $\sim 10^6$ A cm^{-2}, which is still a small increase compared to the observations of heavy ion irradiation in HTSC. We observed maximum enhancement in J_c for H parallel to the defects. The directional pinning enhancement becomes more isotropic at low temperatures, but without an actual crossover to isotropic pinning, which is similar to the effect of columnar defects in YBCO. The stronger pinning by heavy ion tracks observed at the higher temperatures is accompanied by a suppression of the creep rates for fields below the matching field of 1 T. Similar improvement in flux pinning was observed with neutron irradiation in polycrystalline $SmFeAsO_{1-x}F_x$ where the defects are point-like. The relatively small pinning enhancement by columnar defects so far reported in oxypnictides compared to HTSC perhaps reflects higher levels of pinning defects in as-grown samples of the former as compared with the latter materials, but it deserves further detailed exploration.

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