Influence of disorder on the superconducting properties of polycrystalline MgB$_2$

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Abstract. The influence of disorder introduced by neutron irradiation is investigated for sintered bulk material and for in-situ iron sheathed wires. We find strong enhancements of the upper critical field and a decrease of the upper critical field anisotropy. Accordingly, the irreversibility fields, defined as the fields, where the first continuous superconducting current path occurs and the resistivity disappears, are enhanced. The critical current densities increase after irradiation, especially at high magnetic fields. This enhancement is mainly caused by the changes of the reversible properties, but slight changes in flux pinning are observed as well.

1. Introduction

Although grain boundaries do not seem to impede the current flow in polycrystalline MgB$_2$ [1], the granularity of sintered materials strongly affects the field dependence of the critical currents [2]. Due to the $B_{c2}$ anisotropy in MgB$_2$, different grains of a sintered specimen behave differently in magnetic fields due to their different crystallographic orientation with respect to the applied field. Especially grains whose boron planes are oriented perpendicular to the applied field, strongly limit the current flow at high magnetic fields. Thus, for applications which require high current densities, MgB$_2$ can only be used up to the upper critical field of grains with such orientation [3]. A possibility to enhance $B_{c2}$ and to reduce its anisotropy lies in the introduction of disorder, for instance by doping or by irradiation.

2. Experiment

Two small samples were cut out of a large pellet prepared by the HIP process [4]. The resistivity of one sample ($1.5 \times 2.5 \times 11.4$ mm$^3$) was measured as a function of temperature at fixed external field with an applied current of 30 mA. The upper critical field was defined as the applied field at a certain temperature, where the resistivity becomes 90% of its value just above the superconducting transition. The offset ("irreversibility line") was defined by a 10% criterion. Ac susceptibility measurements were performed on a second sample ($2.65 \times 2.1 \times 3$ mm$^3$). At fixed magnetic dc field and at fixed temperature the amplitude of the superimposed ac field was changed stepwise from 0.1 to 10 mT. The measured flux penetration was compared to the
wire

unirr.

$10^{22}$ m$^{-2}$

$2 \times 10^{22}$ m$^{-2}$

$3 \times 10^{22}$ m$^{-2}$

Figure 1. Upper critical field (left) and irreversibility field (right) as a function of temperature for various neutron fluences. The open symbols refer to the bulk sample, the solid symbols refer to the wire.

theoretically expected flux penetration, in order to evaluate $J_c$. The ”theoretical” behavior was calculated numerically based on the Bean model and the actual sample geometry.

Direct transport measurements were performed on a 2.7 mm long piece of an iron sheathed wire, prepared by an in-situ process [5]. $J_c$ was evaluated with a 1 $\mu$V/cm criterion. The superconducting transition at various applied fields was measured in the same way as for the bulk material.

All samples were sequentially irradiated in the central irradiation facility of the TRIGA reactor in Vienna. A cadmium shield was used in order to reduce inhomogeneities in the resulting defect structure. All given neutron fluences refer to fast neutrons (E $> 0.1$ MeV).

3. Results and Discussion

The neutron induced defects are scattering centers for the charge carriers. We find an increase of the normal state resistivity at 40 K from 3.75 $\mu$Ωcm in the unirradiated bulk sample to 12.6, 16.9, 22.2 and 29 $\mu$Ωcm after irradiation to 1, 1.5, 2 and $3 \times 10^{22}$ m$^{-2}$, respectively. While the residual resistivity increases approximately linearly with neutron fluence, the contribution of the phonons remains nearly constant: $\Delta \rho := \rho(300 \text{K}) - \rho(40 \text{K})$ changes from 11.25 $\mu$Ωcm (unirradiated sample) to 12.6 $\mu$Ωcm ($3 \times 10^{22}$ m$^{-2}$). Similar absolute values and a weak change of $\Delta \rho$ with fluence were also reported in Ref. [6] and agree with theoretical calculations, if the conduction is dominated by the $\pi$-bands [7], as expected for clean samples. The normal state resistivity of the superconducting filament of the wire is larger. At 40 K we found $\rho = 160 \mu$Ωcm and $\Delta \rho = 125 \mu$Ωcm. $\Delta \rho$ is larger than expected for dirty samples, where the conductivity is dominated by the $\sigma$-bands [7]. Although $\rho$ can be strongly influenced by voids and/or secondary phases [8] or by a contribution of the grain boundaries, the huge differences in $\rho$ and $\Delta \rho$ still have to be clarified. Unfortunately, no resistivity data are available for the wire’s filament after irradiation, since the iron sheath could not be removed due to its residual radioactivity. The transition temperature of the bulk sample was 38.9 K before the irradiation and decreased almost linearly with increasing neutron fluence to 36.25, 35.33, 34.3 and 32.7 K for 1, 1.5, 2 and $3 \times 10^{22}$ m$^{-2}$. $T_c$ of the wire was significantly smaller (36.7 K), but its change after neutron irradiation was nearly identical: 34.25 and 32.3 K for 1 and $2 \times 10^{22}$ m$^{-2}$. Such a decrease is expected from interband scattering in two band superconductors, but changes of the anisotropy, of the unit cell [6] and of the charge carrier density have also to be taken into account.

The introduction of defects increases the upper critical field, as can be seen in Figure 1.
linear extrapolation to zero temperature, $B_{c2}$ of the bulk sample is estimated to be about 20 T. After the first irradiation it is enhanced to about 32 T, reaching its maximum (∼34 T) at around $2 \times 10^{22}$ m$^{-2}$. Linear extrapolation is quite an inaccurate method to estimate $B_{c2}$ at 0 K. Especially for MgB$_2$, it is a priori unclear whether the slope of the temperature dependence of $B_{c2}$ increases or decreases at low temperatures [9, 10]. Nevertheless, $B_{c2}(0)$ is obviously strongly enhanced after the first irradiation and does not change very much after the following irradiation steps. This is a consequence of two competing effects: the increase of $|\partial B_{c2}/\partial T|$ and the decrease of the transition temperature. We observe a direct correlation between $\rho$, $T_c$ and $B_{c2}$, as predicted for the dirty limit of single band superconductors [11]. Since $\rho$ seems to be dominated by the charge carriers of the $\pi$-bands, the influence of the radiation induced defects on the charge carriers of $\sigma$-bands remains unclear, but disorder was predicted to induce scattering mostly in the $\pi$-bands [7]. In that case, intraband scattering in the $\pi$-bands would be responsible for the enhancement of $B_{c2}$.

As a consequence of the increase in $B_{c2}$, we find a shift of the irreversibility lines [2, 3] (Fig. 1). The upper critical field (∼30.5 T) and the irreversibility fields of the wire (Fig. 2) are significantly higher than those of the bulk specimen in the unirradiated state. This can also be understood in principle by scattering processes, as indicated by the large normal state resistivity. After neutron irradiation, the irreversibility lines of the wire and the bulk specimen become very similar. Although no $B_{c2}$ data of the wire are available after irradiation, because the iron sheath would have to be removed [3], the smaller changes of the irreversibility line indicate that also $B_{c2}$ of the wire increases less than that of the "clean" bulk sample.

The critical current densities of the bulk sample are plotted as a function of the applied field at various temperatures in Fig. 3. The strong field dependence of $J_c$ is a consequence of the rather small $B_{c2}$ in this "clean" sample. The solid lines in Fig. 3 are fits to a percolation model [2]. At low temperatures we find a percolation threshold of 0.2 (which was fixed to this value for all calculations) and an anisotropy of 4.2, which reasonably agrees to results obtained on single crystals [14]. After the irradiation $J_c$ is strongly enhanced at low temperatures and at high fields (Fig. 4). The fits become significantly worse in the irradiated state, which can be explained by the addition of new pinning centers [12], with a different field dependence of $J_c$ (the model is based on grain boundary pinning). The calculated currents underestimate the measured values particularly at intermediate fields, where the radiation induced defects enhance $J_c$, the most in single crystals [13], leading to a pronounced fishtail effect. Nevertheless, the strongly reduced field dependence of $J_c$ is mainly a consequence of the increase in $B_{c2}$ and of a
Figure 4. Influence of the neutron irradiation on the critical current densities. Solid lines are calculation according to a percolation model [2]. Left panel: 5 K (bulk), 4.2 K (wire). Right panel: 20 K.

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decrease in the anisotropy (to ∼3.5 at 2×10^{22} m^{-2}), which was also found in single crystals [14]. The critical currents of the wire behave similarly despite its higher values in the unirradiated state. The enhancements are smaller, but qualitatively the same. At 20 K the beneficial effect of the irradiation is smaller and we even find a degradation of $J_c$ between the first and the second irradiation step in both samples. This is a consequence of the decrease of $T_c$ and the resulting decrease of the pinning energy and of $B_{c2}$ at 20 K (Fig. 1).

4. Conclusion

Neutron irradiation introduces defects in MgB$_2$, which act as scattering centers for the charge carriers. The mean free path of the charge carriers is reduced (the normal state resistivity increases) and the upper critical field is enhanced. The increase of $B_{c2}$ reduces the field dependence of the critical current densities. Indications for additional pinning were found as well. The normal state resistivity, $B_{c2}$ and $J_c$ of the bulk sample and of the wire strongly differ before irradiation, but $B_{c2}$ and $J_c$ become similar after neutron irradiation.

References

[1] Kambara M, Babu N H, Sadki E S, Cooper J R, Minami H, Cardwell D A, Campbell A M and Inoue I H 2001 Supercond. Sci. Technol. 14 L5
[2] Eisterer M, Zehetmayer M and Weber H W 2003 Phys. Rev. Lett. 90 247002
[3] Eisterer M, Krutzler C and Weber H W 2003 J. Appl. Phys. 98 033906
[4] Freidler N A, Li S, Maple M B, Nesterenko V F and Indrakanti S S 2001 Physica C 363 1
[5] Goldacker W, Schlachter S I, Obst B, Liu B, Reiner J and Zimmer S 2004 Supercond. Sci. Technol. 17, S363
[6] Putti M, Braccini V, Ferdeghini C, Gatti F, Grasso G, Manfrinetti P, Marr D, Palenzona A, Pallecchi I, Tarantini C, Sheikin I, Aebersold H U and Lehmam E 2005 Appl. Phys. Lett. 86, 112503 (2005)
[7] Mazin I I, Andersen O K, Jepsen O, Dolgov O V, Kurtus J, Golubov A A, Kuzmenko A B and van der Marel D 2002 Phys. Rev. Lett. 89 107002
[8] Rowell J M 2003 Supercond. Sci. Technol. 16 R17
[9] Gurevich A 2003 Phys. Rev. B 67 184515
[10] Morsor M and Carbotte J P 2005 Phys. Rev. B 72 024538
[11] Orlando T P, McNeill Jr E J, Foner S and Beasley M R 1979 Phys. Rev. B 19 4545
[12] Pallecchi I, Tarantini C, Aebersold H U, Braccini V, Fanciulli C, Ferdeghini C, Gatti F, Lehmam E, Manfrinetti P, Marr D, Palenzona A, Siri A S, Vignolo M and Putti M 2005 Phys. Rev. B 71 212507
[13] Zehetmayer M, Eisterer M, Jun J, Kazakov S M, Karpinski J, Birajdar B, Eibl O and Weber H W 2004 Phys. Rev. B 69 054510
[14] Zehetmayer M, Eisterer M, Jun J, Kazakov S M, Karpinski J and Weber H W 2004 Phys. Rev. B 70, 214516