Flux Pinning in Neutron Irradiated MgB$_2$ Single Crystals.

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Abstract. We report on the effects of neutron irradiation on the irreversible magnetic properties of MgB$_2$ single crystals. The size of the newly created defects is comparable to the superconducting coherence length of MgB$_2$, which makes these defects particularly suitable for pinning flux lines. Indeed, we observe significant quantitative and qualitative modifications of the critical current density. In particular, a second peak (fishtail effects) emerges in the field dependence and is accompanied by history effects. The fishtail effect is thoroughly studied with respect to different neutron fluences and good agreement with a recent theoretical explanation by an order-disorder transition is obtained.

1. Introduction
Neutron irradiation is a well known tool in superconductivity for introducing defects in a more controllable way than by most other methods. In many materials, the new defects are very effective pinning centers, since their size is comparable to the superconducting coherence length and since they are homogeneously distributed. Accordingly, the critical current density is enhanced, which often serves as a benchmark test for other forms of the material modified in different ways. Further, the defect density usually varies linearly with fluence, i.e. the irradiation time.

Recently, we reported on the emergence of a second peak in the field dependence of the magnetic moment in neutron irradiated MgB$_2$ single crystals [1] (for a review on MgB$_2$, see Ref. [2] and references therein). This effect is highly desirable, since it makes $J_c$ quite high at higher fields, but also gives us the opportunity of studying the vortex phase diagram with respect to the defect density. In this article, we review our previous results on $J_c$ and the fishtail effect from Ref. [1] and add new data, mainly in the low fluence range. The fields characterising the fishtail effect are compared with recent theoretical work and discussed in more detail.

2. Experiment
Five single crystals [3] - M1, M2, M3, M4, and M5 - were investigated by different methods before and after neutron irradiation. Their sizes vary between about 350 and 1000 $\mu$m in the basal plane (the ab plane, parallel to the boron planes), and about 20 - 130 $\mu$m along the c-direction. For more experimental details, see Ref. [1] and references therein.
3. Neutron irradiation
The neutron irradiation took place in the central irradiation facility of the TRIGA-MARK-II research reactor in Vienna. In MgB$_2$, neutrons do not only collide with lattice atoms, but are also captured by $^{10}$B, which subsequently decays into $^7$Li and $^4$He [4]. The cross section of the latter reaction is particularly large for thermal neutrons. Thus, the low energy neutrons were shielded by a cadmium foil, in order to avoid inhomogeneities in the defect distribution. The mean free path of the remaining fast neutrons is long enough to ensure a homogeneous defect distribution.

The neutron induced defects were recently observed by transmission electron microscopy [1]. Strain fields with a size of about 10 nm were found, which are typical of radiation induced defects. More detailed investigations on the nature and the density of these defects are currently under way. Nevertheless, the size of these strain fields is comparable to the superconducting coherence length of MgB$_2$. We, therefore, expect significant effects on its irreversible properties.

4. Results and discussion
All crystals have similar reversible properties in the unirradiated state [5, 6], e.g. $T_c \approx 38$–38.4 K, $B_{c2}(0 \text{ K}) \approx 2.7$–3.2 T (for fields $\parallel c$), and a $B_{c2}$ anisotropy of about 4.6 at low temperatures. Somewhat larger differences were found for the irreversible properties, such as $J_c$ and the irreversibility field, which are generally quite small in all samples. Several samples were irradiated sequentially: M1 to an overall fast neutron ($E > 0.1 \text{ MeV}$) fluence ($F_n$) of 1, 2, and $4 \times 10^{20} \text{ m}^{-2}$; M2 to 2, 4, and $6 \times 10^{21} \text{ m}^{-2}$; M3 to $1 \times 10^{22} \text{ m}^{-2}$, and M5 to $5 \times 10^{19} \text{ m}^{-2}$. After irradiation, the width of the transition interval at $T_c$ remains small (< 0.5 K), which indicates a homogeneous defect distribution. The transition temperature decreases almost linearly in the investigated fluence range by about 0.4 K per $10^{21} \text{ m}^{-2}$, whereas $B_{c2}$ increases linearly at the same time by roughly 0.4 T per $10^{21} \text{ m}^{-2}$, i.e. $T_c \approx 34$ K and $B_{c2}(0) \approx 6.9$ T at $F_n = 10^{22} \text{ m}^{-2}$.

Figure 1 presents the most dramatic effects of neutron irradiation on the irreversible properties of MgB$_2$. To reduce the influence of $T_c$ changes, all measurement refer to the same reduced temperature ($T/T_c$) with respect to the unirradiated sample. Generally, neutron irradiation affects $J_c$ by modifying both the defect structure and the reversible properties. For instance, the larger $H_{c2}$ corresponds to a smaller coherence length, but also the thermodynamic critical field is slightly reduced, thus the pinning energy decreases. On the other hand, the defect density...
Figure 2. $H_{od}$ as a function of temperature and neutron fluence (a) and history effects obtained from field cooled measurement (b).

increases leading to higher $J_c$'s. The inset of Fig. 1b shows the fluence dependence of $J_c/J_{c,unir}$ at 5 K and low inductions ($B/B_{c2} = 0.02$) which has a peak at $F_n \sim 2 \times 10^{21} \text{ m}^{-2}$. Similar curves are obtained at higher fields (e.g. $B/B_{c2} = 0.5$) and higher temperatures, but in both cases, the peak is slightly shifted to higher fluences. We conclude, that at low fluences $J_c$ is increased due to a larger defect density, but the modifications of the reversible properties become more and more relevant with increasing fluence.

The most remarkable qualitative effect of neutron irradiation is the emergence of a fishtail (second peak), which could not be detected in any unirradiated sample. This feature is commonly explained by an order-disorder transition of the vortex matter at the field $H_{od}$ (Ref. [7] and references therein). In brief: at fields below $H_{od}$, the vortex-vortex repulsion mainly determines the vortex positions, leading to a highly ordered lattice, which is badly adjusted to the defect matrix, and accordingly $J_c$ is low. Above $H_{od}$ the pinning energy dominates, i.e. the vortices are well adjusted to the defects, $J_c$ is high, but the lattice is rather disordered.

It was argued that the order-disorder transition should refer to a field $H_k$, which is between the onset of the fishtail and the peak, where the slope of the magnetisation changes rather abruptly (cf. Fig. 2b). Experimental results on $H_k$ at low and high $F_n$ are presented in Fig. 2a (symbols). The solid lines refer to theoretical results for $H_{od}$ calculated within the collective pinning theory according to Ref. [7]. Here, $H_{od}$ depends on some experimentally accessible superconducting parameters ($H_{c2}$ and $\gamma$ [5]), the defect radius (which is set to 5 nm), and the unknown defect density ($\rho$), used as the only fit parameter. Experimental and theoretical results agree quite well at low fluences when assuming $\delta L$ pinning (or core pinning, which leads to the same temperature dependence). Here, the calculated fields refer to the bundle regime (in terms of the collective pinning theory) leading to a monotonous decrease of $H_{od}$ with $T$. At high fluences, $H_k$ is found to be almost constant between 0 K and rather high temperatures ($T/T_c \sim 0.5 - 0.8$). For the calculations, we apply the correct $H_{c2}(t)$ ($t = T/T_c$), which follows approximately $(1 - t^{3/2})$ in all our samples, whereas $H_{c2}(t) \propto (1 - t^2)$ - as suggested in Ref. [7] - was used in our previous work. The agreement with experiment is now somewhat worse, but still clearly in favor of $\delta L$ pinning. Even better agreement can be achieved by slightly modifying some parameters in the theory, but we use $c_L = 0.25$ (Lindeman number) and $\zeta = 0.6$ (roughness exponent for a flux line) as in Ref. [7]. The constant behaviour over a large temperature interval refers to the single-vortex pinning regime in the calculations. Actually, the theoretical curve slightly increases in
this regime - not confirmed by the experiment - but this could be eliminated by reducing ζ. At high temperatures, \( H_{od} \) enters the bundle regime at \( H_{sv} \), indicated by arrows in Fig. 2a, which can however be shifted by modifying \( c_L (\sim 0.2 - 0.4) \) to improve the agreement at higher temperatures. For comparison, two curves representing \( \delta T_c \) pinning are also included to Fig. 2a, which clearly do not match any of the experimental curves.

As already mentioned, \( \rho \) was used as a fit-parameter, but actually \( \rho \propto F_n \) is expected. The inset of Fig. 2a presents the experimental \( H_{od}(0 \text{K}) \) vs. \( F_n \) compared with the theoretical results for fixed \( \rho/F_n = 70 \text{ m}^{-1} \). Good agreement is found at low fluences, whereas the theoretical curves overestimate the effect of the defect density at high fluences. Still the overall agreement is satisfactory. Differences may be explained by insufficiencies of the collective pinning theory, since the defects are rather large, but the radius of about 5 nm is still smaller than the coherence length of 11 - 7 nm at 0K, which is often used to justify the applicability of this theory.

Finally, we briefly discuss the history effects, which are considered to be a strong argument for the order-disorder transition. History effects are commonly observed close to a phase transition in physical systems due to “overheating” or “supercooling” of one phase. We assume that the amount of disordered phase in the superconductor is larger below \( H_{od} \) in the decreasing field branch than in the increasing one - corresponding to a larger \( J_c \) there (note the very different fields of the onset in increasing and the offset of the fishtail in decreasing fields already seen in the major loop of Fig. 2b). Large history effects from minor hysteresis loops were reported in Ref. [1]. Here, we concentrate on the effects found from field-cooled measurements shown in Fig. 2b. The sample is successively cooled down at various fixed fields (so that, e.g., point A - inset of Fig. 2b - is reached), followed by a measurement at increasing or decreasing fields. Fig. 2b compares such curves with the major loop. By field cooling, particularly large disorder is induced in vortex matter, accordingly the (absolute value of the) irreversible magnetic moment is larger than in the major loop after some field changes. However, this holds only for fields below \( H_k \), supporting \( H_k = H_{od} \), and the differences are larger for increasing fields, supporting the assumption that the amount of disorder is smaller in the increasing field branch of the major loop than in the decreasing one.

In summary, we found that the critical current density increases with neutron irradiation at low fluences due to the larger defect density, but decreases at higher fluences because of a modification of some superconducting parameters. The second peak emerging in \( J_c(B) \) after neutron irradiation and the accompanying history effects were studied in detail. They are found to be well described by order-disorder theory of vortex matter.

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References

[1] 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
[2] For a review, see Physica C 385 (1-2) (2003).
[3] Karpinski J, Angst M, Jun J, Kazakov S M, Puzniak R, Wisniewski A, Roos J, Keller H, Perucchi A, Degiorgi L, Eskildsen M, Bordet P, Vinnikov L and Mironov A 2003 Supercond. Sci. Technol. 16, 221
[4] Eisterer M, Zehetmayer M, Toennis S, Weber H W, Kambara M, Hari Babu N, Cardwell D A and Greenwood L R 2002 Supercond. Sci. Technol. 15, L9
[5] Zehetmayer M, Eisterer M, Jun J, Kazakov S M, Karpinski J and Weber H W 2004 Phys. Rev. B 70, 214516
[6] Zehetmayer M, Eisterer M, Weber H W, Jun J, Kazakov S M, Karpinski J and Wisniewski A 2002 Phys. Rev. B 66, 052505
[7] Mikitik G and Brandt E H 2001 Phys. Rev. B 64, 184514