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LETTER TO THE EDITOR

ENHANCEMENT OF FLUX PINNING IN NEUTRON IRRADIATED MgB_2 SUPERCONDUCTOR

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$m - H$ loops for virgin and neutron irradiated bulk and powder samples of MgB_2 were measured in the temperature range $5 - 30$ K in magnetic field $B \leq 1$ T. The irradiation at thermal neutron fluences $9 \cdot 10^{13}$ and $4.5 \cdot 10^{14}$ cm^{-2} caused very small enhancement of $m - H$ loops at lower temperatures ($T < 20$ K), whereas the effect at high temperatures was unclear due to the difficulty in achieving exactly the same measurement temperature prior and after irradiation. However, the irradiation at $4.5 \cdot 10^{15}$ cm^{-2} produced clear enhancement of $m - H$ loops (hence J_c) at all investigated temperatures, which provides the evidence for the enhancement of flux pinning in MgB_2 due to ion tracks resulting from n+^{10}B reaction. The potential of this technique for the enhancement of flux pinning in high temperature superconductors is briefly discussed.

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1. *Introduction*

The discovery of new superconductor [1] MgB_2 aroused a large interest in the scientific community [2]. Compared to high temperature superconductors (HTS), MgB_2 has lower transition temperature, $T_{co} \cong 39$ K, but its simple composition, abundance of constituents and probable absence of the problems associated with weak intergranular couplings [3,4] (inherent to HTS) make MgB_2 promising material for applications in the temperature range $20-30$ K which is still well above T_cs for low temperature superconductors (LTS). However, compared to LTS employed in the applications of superconductivity, MgB_2 shows weaker flux pinning which manifests itself in sizably lower critical current density [5] J_c at 4.2 K. Moreover, flux pinning in MgB_2 decreases rapidly at elevated temperatures, rendering quite low [6] J_c for $T \geq 20$ K. Since as of yet very little effort has been put into optimizing J_c of MgB_2, there is ample space for the improvement of flux pinning in MgB_2.

In this respect, MgB_2 presents almost ideal testground for testing our earlier proposal [7] that flux pinning in boron doped HTS and borocarbides [8] can be enhanced via ion tracks associated with the n + ^{10}B reaction. For these purposes in the case of MgB_2 one can use ^{4}He and ^{7}Li ions from ^{10}B(n,α)^{7}Li reaction. Due to its large cross section at thermal energies ($\sigma_0 = 3837 \times 10^{-28}$ m^2) and high abundance of ^{10}B in natural boron (19.78%), one can reach high density of ion tracks in MgB_2 sample exposed to modest thermal neutron fluences. (This contrasts sharply with U/n and Bi/p treatments employed for the increase of flux pinning in HTS which require high fluences of thermal neutrons and high energy protons, respectively [9].) At thermal energies, the reaction proceedes only via ^{7}Li ground and first excited states with the α_0/α_1 branching of 6.7%, i.e. the main contribution comes from the reaction leading to the excited state. In this case, the reaction products ^{4}He and ^{7}Li nuclei have energies of 1.47 and 0.84 MeV, respectively. The sum of their ranges in MgB_2 with $\rho = 2.6$ gcm^{-3} is about 6 μm.

Large cross section σ_0 for ^{10}B(n,α)^{7}Li reaction and high boron content in MgB_2 poses problems in irradiation of bulk samples. The mean free path of thermal neutrons in MgB_2 is about 0.2 mm. Obviously, in samples with thickness of ~ 1 mm the ion tracks will be very unevenly distributed. In an isotropic neutron field, more than half of the ion tracks would be in the first 100 μm from the surface. The problem is somewhat facilitated in thinner samples, but the homogenous distribution of ion tracks in the samples is still unlikely.

The simplest way to monitor the enhancement of flux pinning (an increase in J_c) in type II superconductors is to measure the magnetization hysteresis curves ($m-H$ loops). Since J_c is proportional to irreversible magnetization, any increase in the breadth of $m-H$ loop shows the enhancement of flux pinning. This method can also be applied to powder samples where more direct transport measurements of J_c are not possible. In what follows, we present the preliminary results of a magnetization study performed on virgin and neutron irradiated bulk and powder samples of MgB_2. Although these results were obtained for rather thick samples ($d \geq 1$ mm) and quite low neutron fluences, they indicate an enhancement of flux pinning following neutron irradiation both in bulk and powder samples. Bulk MgB_2 samples were cut from a pellet prepared by conventional solid state reaction [10]. SEM studies revealed coarse-grained structure with grain size of the order of 200 μm [10]. The measured density was ~ 1 gcm^{-3}, i.e. about 0.4 of the ideal density of MgB_2. The sample for magnetization study had dimensions 3.8 mm $\times$ 1.5 mm $\times$ 1 mm and a mass of 5.93 mg. A similar sample (cut from the adjoining part of the pellet) was used for ac susceptibility study of superconducting transition temperatures prior and after neutron irradiation. The powder sample was prepared by mixing small amount of commercial (Alfa Aesar) MgB_2 powder (325 MESH size) with about three times larger volume of epoxy in a plastic pill. After setting, the plastic container was removed and a cylindrical sample with diameter 5.2 mm and length about 5 mm was split into two approximately semicylindrical samples used for magnetization and ac susceptibility measurements, respectively. The $m - H$ loops were measured with commercial vibrating sample magnetometer (VSM) in the magnetic field up to 1 T at the field sweep rate of 15 mT/s. Ac susceptibility was measured with a highly sensitive ac susceptometer [11]. The employed temperature range was $5 - 40$ K. The samples were irradiated at the roundabout of the TRIGA Mark II reactor of the Jožef Stefan Institute in Ljubljana. At the reactor power of 25 kW, the rotational irradiation facility has a flux of $1.45 \cdot 10^{11}$ cm^{-2}s^{-1} thermal ($E < 0.5$ eV) and $0.22 \cdot 10^{11}$ cm^{-2}s^{-1} fast ($E > 0.1$ MeV) neutrons. The initial thermal neutron fluence for all samples was $9 \cdot 10^{13}$ cm^{-2}. In later experiments, we applied five and fifty times larger thermal neutron fluences.

The ac susceptibility of the bulk virgin sample (Fig. 1) showed the superconducting transition with the diamagnetic onset at $T_{\rm co} = 38.2$ K and a transition width $\Delta T_c(0.1 - 0.9) = 0.46$ K in the ac field of amplitude 1.5 A/m. In spite of its porosity, sample showed excellent grain connectivity as manifested by smooth, single-step transitions at elevated ac fields ($\sim 10^4$ A/m). The comparison of low-field – low-temperature diamagnetism in our sample with that in niobium sample of approximately the same shape indicated the Meissner fraction $\geq 80\%$. The powder sample (Fig. 1) showed a similar $T_{\rm co} = 38.2$ K (inset to Fig. 1), but most of the

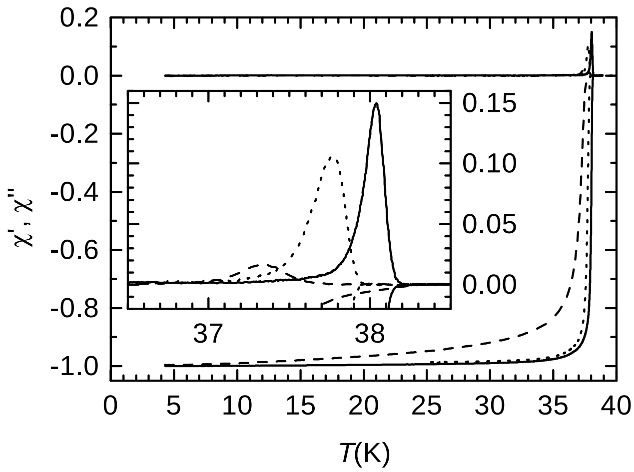

Fig. 1. Variations of real (χ') and imaginary (χ'') part of ac susceptibility for commercial MgB_2 powder embedded in epoxy (dashed) and bulk MgB_2 sample prior (full) and after irradiation at thermal neutron fluence $4.5 \cdot 10^{15}$ cm^{-2} (dotted) with temperature for alternating field amplitude $H_m = 15.8$ A/m. The inset: enlarged view of the onset of superconductivity for the same samples.

transition was shifted to lower temperature compared to that of the bulk sample. At low field (1.5 A/m), this shift was 0.9 K at half transition, and the transition showed a shallow tail bellow 35 K which persisted down to 4.2 K (the transition in bulk sample was completed at 35 K). These phenomena are a consequence of a broad grain size distribution (grain sizes ≤ 43 μm) and impurity content in commercial MgB_2 powder. Both for powder and bulk samples, the superconducting transitions, after irradiations to $9 \cdot 10^{13}$ and $4.5 \cdot 10^{14}$ cm^{-2}, remained practically the same as those for virgin samples. In particular, T_{co} remained within 0.1 K of the initial value and the breadths and magnitudes of the diamagnetic transitions practically did not change. However, the neutron fluence of $4.5 \cdot 10^{15}$ cm^{-2} caused a small shift of the diamagnetic transition (≈ 0.3 K at half transition) towards lower temperature for the bulk sample (Fig. 1).

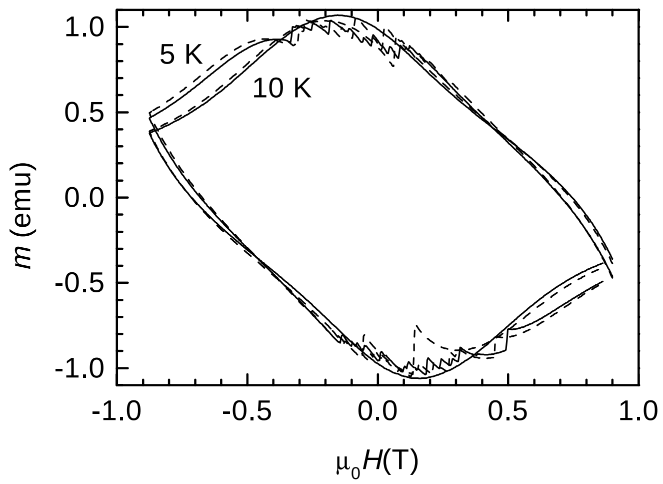

Fig. 2. Magnetization loops for bulk MgB_2 sample at 5 and 10 K. Dashed line denotes results obtained after irradiation of sample to thermal neutron fluence of $9 \cdot 10^{13}$ cm^{-2}.

Figure 2 shows the dependence of the magnetic moment vs. the applied field loops for the bulk sample at 5 and 10 K. The results for both virgin and irradiated (dashed line) sample are shown. The field sweep rate was 15 mT/s. At elevated fields, the irradiation seems to produce marginal increase in the irreversible magnetic moment, and this increase is somewhat more pronounced at 10 K than at 5 K. This result is consistent with the rather low applied neutron fluence, uneven distribution of ion tracks in our thick sample (most tracks were concentrated within thin surface layers) and much stronger (dominant) pinning effect of intrinsic pinning centres which manifests itself in high critical current densities of the bulk MgB_2 samples at low temperatures [5].

However, the reappearance of the partial flux jumps (manifested as sharp roughly zig-zag variation of magnetic moment at lower field in Fig. 2) in irradiated sample at 10 K (note that virgin sample did not show flux jumps at 10 K) seems to support the enhancement of flux pinning at 10 K after neutron irradiation. These partial (pseudo) flux jumps have already been reported for bulk MgB_2 samples similar to our results [10] at low temperatures ($T < 10$ K). In contrast to the normal flux jumps in LTS [12], the above phenomenon is associated with a violent flux entry into the surface layers of porous sample. The phenomenon occurs only at low temperatures when intragranular flux pinning greatly exceeds that in intergranular links. Therefore, the reappearance of this phenomenon upon irradiation seems to indicate an increase in the intragranular pinning associated with ion tracks.

The $m-H$ loops for both virgin and neutron irradiated ($9 \cdot 10^{13}$ cm^{-2}) powder sample were measured at temperatures 5, 10, 20 and 30 K. For $T = 5$ and 10 K, the $m - H$ loops of irradiated powder were identical to those of the virgin one, whereas at 20 and 30 K $m-H$ the loops of the irradiated sample showed small and sizable enhancement, respectively. No effect of light irradiation on $m - H$ loops of MgB_2 powder for $T \leq 10$ K was plausible. It probably arised from a combination of strong (dominant) intrinsic flux pinning in MgB_2 powders (manifested by an order of magnitude larger J_cs of MgB_2 powders compared to those of corresponding bulk samples [3,13]) and the applied low neutron fluence (hence low ion track density). However, a large increase in the breadth of $m-H$ loops of irradiated powder at 30 K was unexpected. Although the intrinsic flux pinning in MgB_2 grains decreases rapidly at elevated temperatures [6], the pinning by ion tracks also becomes less efficient due to the corresponding increase of the coherence length of MgB_2 [5] with temperature. Therefore, a large effect of low neutron fluence on $m-H$ loops at 30 K seems unlikely.

Furthermore, rapid variations of J_c with temperature for MgB_2 samples at elevated temperatures [3,5,6,10,13] makes the comparison of high temperature $m-H$ loops obtained in two experiments reliable only if the sample temperatures were practically identical in the two experiments. This condition is difficult to fulfill with the temperature control technique commonly used for VSMs. In VSM, preheated helium gas flows around the sample and there is no thermometer at the sample holder. Therefore, the actual sample temperature depends somewhat on the combination of the helium flow rate, duration of the temperature stabilization and the size and properties (such as shape, thermal capacity and conductivity, etc.) of the sample. Because of this, we performed additional experiments in order to ascertain the role of ion tracks in flux pinning in MgB_2. In the second experiment, we irradiated two powder and one bulk MgB_2 sample to five times larger neutron fluence ($4.5 \cdot 10^{14}$ cm^{-2}) than that used in the first experiment and measured their $m-H$ loops prior and after irradiation at temperatures 10, 20 and 30 K. In order to minimize the possible difference in sample temperature in subsequent measurements, we prepared plate-like powder samples with thickness $d \approx 1$ mm and used longer time intervals for the temperature stabilization ($t_s \geq 15$ min).

Figure 3 shows $m-H$ loops both for virgin and for irradiated ($4.5 \cdot 10^{14}$ cm^{-2}) MgB_2 powder at temperatures 10, 20 and 30 K, respectively. The breadth of $m-H$ loop after irradiation is slightly larger at 10 K, shows almost no change at 20 K and appears somewhat smaller at 30 K. The results with other powder samples were

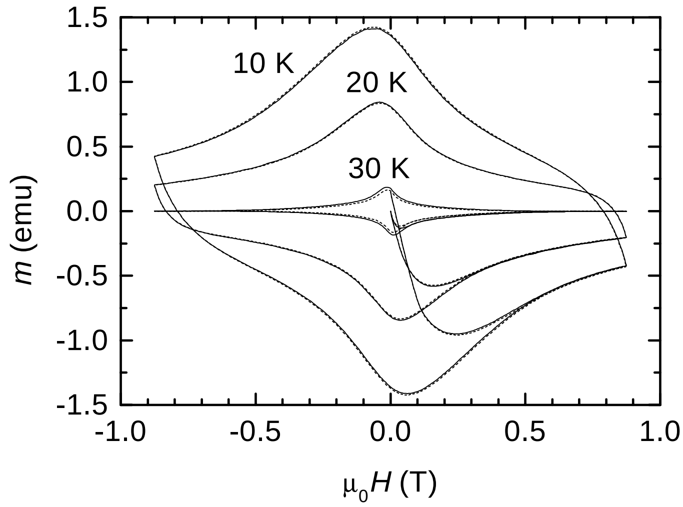

Fig. 3. Magnetization loops for MgB_2 powder embedded in epoxy at 10, 20 and 30 K, respectively. Dashed line is for the same sample exposed to neutron fluence of $4.5 \cdot 10^{14}$ cm^{-2}.

practically the same. Whereas the results for 10 K and 20 K can be interpreted in terms of decreasing efficiency of ion track pinning on increasing temperature, the decrease of m (hence J_c) of irradiated sample at 30 K is at variance with the fact that irradiation produced no measurable change in either transition temperature or the shape of transition for MgB_2 powders. Non-appearance of the change in the superconducting parameters of the samples is difficult to reconcile with the suppression of their J_cs at 30 K. The most probable explanation of this observation (Fig. 3) is that our control of the sample temperature, although improved, is still insufficient for reliable measurements of small changes in J_c at 30 K. The $m - H$ loops of the bulk MgB_2 sample irradiated to the same neutron fluence was at 20 K almost unchanged compared to that for virgin one, whereas at 30 K it showed small enhancement. No enhancement of $m - H$ loop at 20 K probably indicates that the enhancement at 30 K was at least partially due to a slightly lower measurement temperature after irradiation. However, the $m - H$ loops of irradiated sample showed strong flux jumps at 10 K (which were not observed in the virgin sample) thus clearly indicating an enhancement of flux pinning (larger J_c [14]) after irradiation.

Taken together, these experiments show small enhancement of flux pinning at lower temperatures ($T < 20$ K) in irradiated MgB_2 samples (both bulk and powder ones). However, for the applied (low) neutron fluences, the effects are to small to allow more quantitative assessment. Therefore, we performed the third experiment in which two powder samples and one bulk MgB_2 sample were irradiated to neutron fluence of $4.5 \cdot 10^{15}$ cm^{-2}. After irradiation, the superconducting transition of bulk sample was shifted a little towards lower temperature, the shift at half transition being about 0.3 K (Fig. 1). Since the powder samples showed an induced radioactivity, probably caused by some impurity present in commercial MgB_2 powder (95.5% purity), the measurements on these samples were postponed.

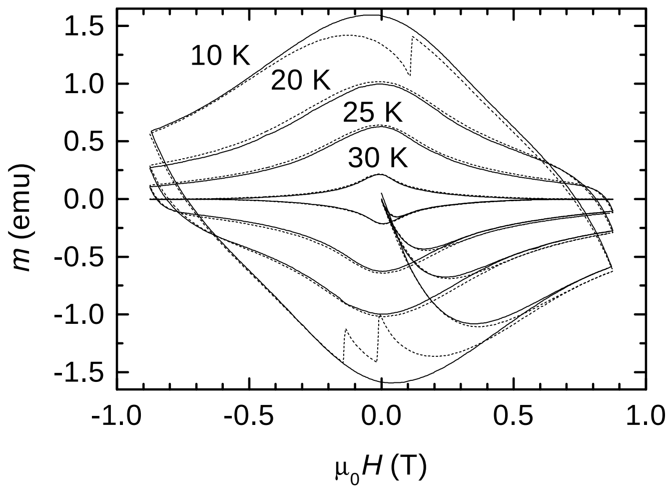

Fig. 4. Magnetization loops for bulk MgB_2 sample at 10, 20, 25 and 30 K, respectively. Dashed line is for the same sample exposed to the neutron fluence $4.5 \cdot 10^{15}$ *cm^{-2}.*

Figure 4 shows $m - H$ loops both for irradiated and virgin bulk MgB_2 sample at temperatures 10, 20, 25 and 30 K, respectively. The appearance of flux jumps at 10 K and the increase in the breadths of $m - H$ loops for irradiated sample at all other temperatures prove the enhancement of the flux pinning after irradiation. We also observe that the enhancement of J_c for $T \geq 25$ K decreases with increasing temperature, which is partially due to the decrease of T_c upon irradiation (Fig. 1). The enhancement of J_c due to ion-track flux pinning at 20 K is about 2% and 5% at $\mu_0 H = 0$ and 0.6 T, respectively. Clearly, the enhancement of J_c will become larger at higher applied fields. The observed rather modest enhancement of J_c in irradiated bulk MgB_2 sample is probably a consequence of a quite large coherence length in MgB_2, small masses and energies of ions and the use of still rather modest neutron fluence. Since, in spite of the porosity of our sample, the decrease of T_c is quite small, the use of sizably larger neutron fluences seems feasible, which would result in correspondingly larger enhancement of flux pinning. Another advantage of flux pinning by ion tracks resulting from n+^{10}B reaction is that J_c is enhanced at all fields (including $H = 0$, Figs. 3 and 4), whereas the other ion tracks invariably suppress J_c at $H = 0$, and the enhancement of J_c occurs only at elevated fields [9]. Furthermore, the enhancement of flux pinning by ion tracks from n+^{10}B reaction in HTS (such as Bi–Sr–Ca–Cu–O compounds) should be sizably larger due to considerably smaller coherence lengths and weaker intrinsic flux pinning in these materials. Taken together, our results confirm the flux pinning effect of ion tracks in MgB_2 and moreover enable us to predict that the same technique may become powerful method for the enhancement of flux pinning in HTS [7].

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References

[1] J. Nagamatsu, N. Nagakawa, T. Murnaka, Y. Zenitani and J. Akimitsu, Nature **410** (2001) 63.

[2] See for instance, Nature **410** (2001) 186.

[3] D. C. Larbalestier et al., Nature **410** (2001) 193.

[4] B. A. Glowacki, M. Majoros, M. Vickers, J. E. Evets, Y. Shi and I. McDougall, Supercond. Sci. Technol. **14** (2001) 193.

[5] P. C. Canfield, D. K. Finnemore, S. L. Bud'ko, J. E. Ostenson, G. Lapertot, C. E. Cunningham and C. Petrovic, Phys. Rev. Lett. **86** (2001) 2423, and ref. therein.

[6] Y. Boguslavsky, G. K. Perkins, X. Qi, L. F. Cohen and A. D. Caplin, Nature **410** (2001) 563.

[7] Đ. Miljanić, (2000) unpublished.

[8] P. C. Canfield, P. L. Gammel and D. J. Bishop, Phys. Today **51** (1998) 40.

[9] E. Babić et al., (2001) submitted for publication.

[10] S. X. Dou, X. L. Wang, J. Horvat, D. Milliken, E. W. Collings and M. D. Sumption, (2001) submitted to Appl. Phys. Lett.

[11] Đ. Drobac and Ž. Marohnić, FIZIKA A (Zagreb) **8** (1999) 165.

[12] P. S. Swartz and C. P. Bean, J. Appl. Phys. **39** (1968) 4991.

[13] M. Kambara et al., Supercond. Sci. Technol. **14** (2001) L5.

[14] S. Jin, H. Mavoori, C. Bower and R. B. van Dover, Nature **411** (2001) 563.

POJAČANJE ZAPINJANJA MAGNETSKIH VRTLOGA U NEUTRONIMA OZRAČENOM SUPRAVODIČU MgB_2

Ozračivanje termičkim neutronima do $9 \cdot 10^{13}$ i $4.5 \cdot 10^{14}$ n/cm^2 uzrokovalo je vrlo malo proširenje $m-H$ krivulja na nižim temperaturama ($T < 20$ K), dok je učinak na višim temperaturama nejasan zbog poteškoće da se postigne točna jednakost mjerne temperature prije i poslije ozračivanja. Ozračivanje na $4.5 \cdot 10^{15}$ n/cm^2 uzrokovalo je jasno proširenje $m-H$ krivulja na svim temperaturama, što ukazuje da ionski tragovi nastali n+^{10}B reakcijom pojačavaju zapinjanje linija magnetskog toka u MgB_2. Kratko se razmatra prikladnost tog postupka za pojačanje zapinjanja linija toka u visokotemperaturnim supravodičima.