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# Uranium Doping and Thermal Neutron Irradiation Flux Pinning Effects in MgB<sub>2</sub>

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# Uranium Doping and Thermal Neutron Irradiation Flux Pinning Effects in MgB<sub>2</sub>

Tania M. Silver, Joseph Horvat, Mark Reinhard, Pei Yao, Shokat Keshavarzi, Paul Munroe, and Shi Xue Dou

**Abstract**—The U/n method is a well-established means of improving flux pinning and critical current performance in cuprate superconductors. The method involves the doping of the superconductor with <sup>235</sup>U followed by irradiation with thermal neutrons to promote fission. The resultant columnar damage tracks produced by the energetic fission products pin flux vortices and improve critical current performance in magnetic fields. No such improvement could be observed when the U/n method was applied to the MgB<sub>2</sub> superconductor. No fission tracks could be observed in TEM, even for samples that were irradiated at the highest fluence. Gamma-ray spectroscopy indicated that fission had occurred in the expected way. The likely resistance of MgB<sub>2</sub> to the formation of fission tracks is highly relevant to attempts to improve flux pinning and superconducting performance in this material through the introduction of columnar defects.

**Index Terms**—Flux pinning, MgB<sub>2</sub>, U/n method, uranium doping.

## I. INTRODUCTION

SINCE its superconducting properties were first discovered in 2001 [1], [2], MgB<sub>2</sub> has been found to share characteristics with both conventional low-temperature superconductors (LTS) and with cuprate high-temperature superconductors (HTS) [3]. Its critical temperature  $T_c$  is nearly 40 K, unusually high for a noncuprate material. Although MgB<sub>2</sub> films have attained critical currents above  $10^6$  A/cm<sup>2</sup> at 4.2 K in low magnetic fields [4], MgB<sub>2</sub> [3] and the HTS cuprates [5] share a rapidly decreasing  $J_c(H)$  performance as magnetic fields increase, compared to LTS superconductors such as Nb<sub>3</sub>Sn and Nb–Ti. In HTS materials, the field performance is related to both weak links between grains and poor flux pinning behavior. The former consideration does not apply to MgB<sub>2</sub>, as transport measurements of  $J_c$  and measurements calculated from magnetic hysteresis loops yield very similar results, indicating that the flow of supercurrent is not hindered by grain boundaries [6],

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[7]. The flux creep in MgB<sub>2</sub> is much weaker than in average HTS [8], [9], indicating stronger vortex pinning. However, this pinning still needs to be improved to offset the effect of the low  $H_{c2}$  of MgB<sub>2</sub> (about 18 T) [10] on the field dependence of  $J_c$ .

Superconductivity above  $H_{c1}$  in any type-II material depends on flux pinning in regions where the superconducting order parameter is reduced, either by intrinsic features of the crystal structure or by point or extended defects. This prevents motion of the flux vortices due to Lorentz forces, resulting in local superconducting phase slip and a nonzero electrical resistance [11]. Strong pinning centers and  $J_c$  improvements in HTS superconductors have been produced over the years by means such as the introduction of precipitates and the use of a variety of irradiation techniques employing neutrons, heavy ions, electrons, and protons [5]. Some methods of introducing pinning centers have also been at least partially successful with MgB<sub>2</sub>, including oxygen alloying in thin films [4] and the addition of nanoscale particles of SiC [12] and Si [13], as well as irradiation with protons [14] and neutrons [15], [16].

Heavy ion irradiation is used to introduce columnar defects into HTS materials. Narayan *et al.* [17] irradiated polycrystalline MgB<sub>2</sub> with 200 MeV <sup>107</sup>Ag ions. The surfaces of the irradiated samples were imaged with scanning tunneling microscopy (STM), and there was evidence of amorphous tracks 65 Å in diameter. The lengths of the tracks could not be determined, but computer modeling indicated that the ions should penetrate 15 μm into the material. They did not attempt to determine  $J_c$  in the irradiated samples. Olsson *et al.* [18], on the other hand, irradiated 4000-Å thick *c*-axis oriented MgB<sub>2</sub> films with 1.2 GeV U<sup>57+</sup> ions and 1.4 GeV Au<sup>32+</sup> ions with the beam parallel to the *c* axis. They also examined the surfaces of their irradiated samples with STM and found no evidence of columnar defects in any of the samples. They also found no significant differences in magnetization and, hence,  $J_c$  between the irradiated and unirradiated samples. Because a number of parameters are involved, as described below, in determining whether amorphous columnar defects will form in a particular material, the question remains open for MgB<sub>2</sub>.

Another similar irradiation method that has not yet been reported for MgB<sub>2</sub> is the U/n [19], [20] method that is the subject of the present work; although, it has been highly successful in improving flux pinning in HTS. The U/n method differs from straightforward irradiation with thermal neutrons in that the material is first doped with compounds containing <sup>235</sup>U. When irradiation takes place the <sup>235</sup>U atoms absorb thermal neutrons and fission. The two fission products recoil in opposite directions with a total kinetic energy of approximately 160 MeV, creating extended defects in HTS in the form of two fission tracks.

Fission tracks are made up of amorphous material and are approximately  $10\ \mu\text{m}$  long and  $10\ \text{nm}$  in diameter in Bi-based HTS superconductors [21]. The track structure is discontinuous and randomly oriented.

U/n achieves its greatest success with highly anisotropic Bi-based (BSCCO) superconductors such as Bi-2223 (anisotropy coefficient  $\gamma = 31$  [22]), where a 500-fold improvement in  $J_c$  was reported for Ag-clad tape [23] at  $77\ \text{K}$  and  $0.8\ \text{T}$  for  $H//c$ . There was also improvement after irradiation for  $H//ab$ , but  $J_c$  improved less than one order of magnitude. The ratio  $J_c(H//ab)/J_c(H//c)$  was reduced 23 times at  $0.5\ \text{T}$  and  $77\ \text{K}$ . A lesser 20-fold improvement in  $J_c$  at  $77\ \text{K}$  and  $0.25\ \text{T}$  has been reported [24] in bulk melt textured  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO), which is much less anisotropic ( $\gamma = 5$  [25]). This is because the vortices in HTS are two-dimensional (2-D) pancake vortices, residing on the  $\text{CuO}_2$  planes [26], [27]. In highly anisotropic HTS, these pancake vortices in each  $\text{CuO}_2$  layer can move independently from the vortices in the other  $\text{CuO}_2$  layers. However, if the pinning centers are in the shape of long columns traversing the  $\text{CuO}_2$  planes, the pancake vortices in different layers will be collectively pinned along these columns and will thus be effectively aligned into vortex lines without an unacceptably high density of defects. Such pinning has been shown to be much more effective than pinning by point-like pinning centers [5].

In  $\text{MgB}_2$ , reported results for the anisotropy coefficient  $\gamma = \xi_{ab}/\xi_c$  range from 1.1 to 2.7 for textured bulk, aligned crystallites, films, and single crystal [3], [28], indicating that enormous improvements cannot be expected from fission tracks or other columnar defects. There is no evidence of a 2-D vortex state in  $\text{MgB}_2$  bulk material, and the I-V characteristics have been reported as consistent with a vortex glass model [29], [30]. However, as with HTS, the reported coherence length values [3] of  $\xi_{ab}(0) = 3.7\text{--}12\ \text{nm}$  and  $\xi_c(0) = 1.6\text{--}3.6\ \text{nm}$  are of the same order as the expected diameter of the fission tracks introduced by high energy fragments resulting from the fission of  $^{235}\text{U}$ . It has been generally accepted that the optimal size of a pinning center is of the order of the coherence length [31].

The most serious problem for the U/n method in  $\text{MgB}_2$  is the enormous cross section of 3837 barn ( $b$ , where  $1b = 10^{-28}\ \text{m}^2$ ) for the  $^{10}\text{B}(n, \alpha)^7\text{Li}$  nuclear reaction at thermal (of the order of 25 meV) neutron energies. Here, a  $^{10}\text{B}$  nucleus and a neutron react to form an  $\alpha$  particle of kinetic energy 1.47 MeV and a  $^7\text{Li}$  nucleus with a kinetic energy of 0.84 MeV. By comparison, the  $^{235}\text{U}$  fission cross section is  $584\ b$ . Natural boron is comprised of 19.9%  $^{10}\text{B}$ , with the balance  $^{11}\text{B}$ . The  $^{10}\text{B}(n, \alpha)^7\text{Li}$  reaction was deliberately used by Babic *et al.* [15] in an attempt to improve flux pinning by introducing ion tracks into  $\text{MgB}_2$ . They reported an enhancement in the upper critical field (as did Eisterer *et al.* [16], who also studied neutron irradiation of  $\text{MgB}_2$ ) and an enhancement of magnetization hysteresis loops and hence  $J_c$ . However, the distribution of defects was extremely inhomogeneous in both reports because the high content of  $^{10}\text{B}$  would have only allowed the neutrons to penetrate into a thin surface layer before being completely absorbed. In using the U/n method, we essentially eliminated the competition for thermal neutrons between  $^{10}\text{B}$  and  $^{235}\text{U}$  by using highly enriched  $^{11}\text{B}$  instead of natural boron. ( $^{11}\text{B}$  has a total thermal neu-

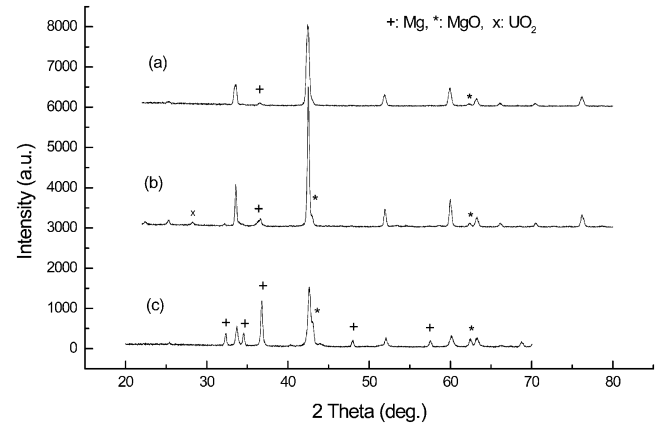


Fig. 1. XRD spectra of the  $\text{MgB}_2$  used for the samples. (a) Undoped, sintered at  $900\ \text{°C}$  for 3 h; (b) doped with 1 wt% U as  $\text{UO}_2$ , sintered at  $900\ \text{°C}$  for 3 h; and (c) undoped, sintered at  $760\ \text{°C}$  for 0.5 h.  $\text{MgB}_2$  phase peaks are unmarked.

tron cross section of  $5.05\ b$ , almost all of it representing elastic scattering.) The negative results we describe below thus strongly support the position of [18] since they cannot be attributed to self-shielding effects.

## II. EXPERIMENT

To test the applicability of the U/n technique for enhancing  $J_c$  in  $\text{MgB}_2$ , an experimental program was designed. Powders of  $1\text{--}11\ \mu\text{m}$  Mg from Hypertech (for samples A-D) or 325 mesh Mg (for sample E) and crystalline B (99.5 at%  $^{11}\text{B}$ , particle size  $<22\ \mu\text{m}$ , from Eagle-Picher) were mixed in a mortar in a stoichiometric atomic ratio of 1:2. Half of the powders were mixed with  $\text{UO}_2$  (93% enriched in  $^{235}\text{U}$ ) to give 1 wt% U. Optical microscopy revealed that the  $\text{UO}_2$  particles ranged from  $2\text{--}4\ \mu\text{m}$  in diameter.

The powders were pressed into 0.4 g pellets and the pellets sealed in iron tubes. Sintering took place in a tube furnace under flowing argon. For samples A-D the temperature was ramped up to  $760\ \text{°C}$  over 1 h. The pellets were held at that temperature for 30 min and then furnace cooled. For sample E, the temperature was ramped up to  $900\ \text{°C}$  over 1.5 h, and the pellets were held at that temperature for 3 h, then furnace cooled. The densities of the samples were  $1.4\ \text{g/cm}^3$  (samples A-D) and  $1.0\text{--}1.1\ \text{g/cm}^3$  (sample E). X-ray diffraction (XRD) was used to determine the phase composition of the doped and undoped samples, while energy dispersive spectroscopy (EDS) was used to determine the uranium distribution of doped samples. Portions of the doped and undoped samples were then cut and filed into small regular rectangular blocks with dimensions of typically  $3\text{--}4 \times 3 \times 2\ \text{mm}^3$ . The magnetization and ac susceptibility were determined using a quantum design physical properties measurement system (PPMS) in a time-varying magnetic field with sweep rate  $50\ \text{Oe/s}$  and amplitude up to  $8.5 \times 10^4\ \text{Oe}$ , with the applied magnetic field parallel to the longest dimension of the sample. The low-field zero field cooled (ZFC) and field cooled (FC) measurements were made with a quantum design magnetic properties measurement system (MPMS). The  $J_c$  values at different magnetic fields and temperatures were calculated from the height  $\Delta M$  of the magnetic hysteresis loops using the Bean model:  $J_c = 20\Delta M/[a(1 - a/3b)]$ , where  $a$

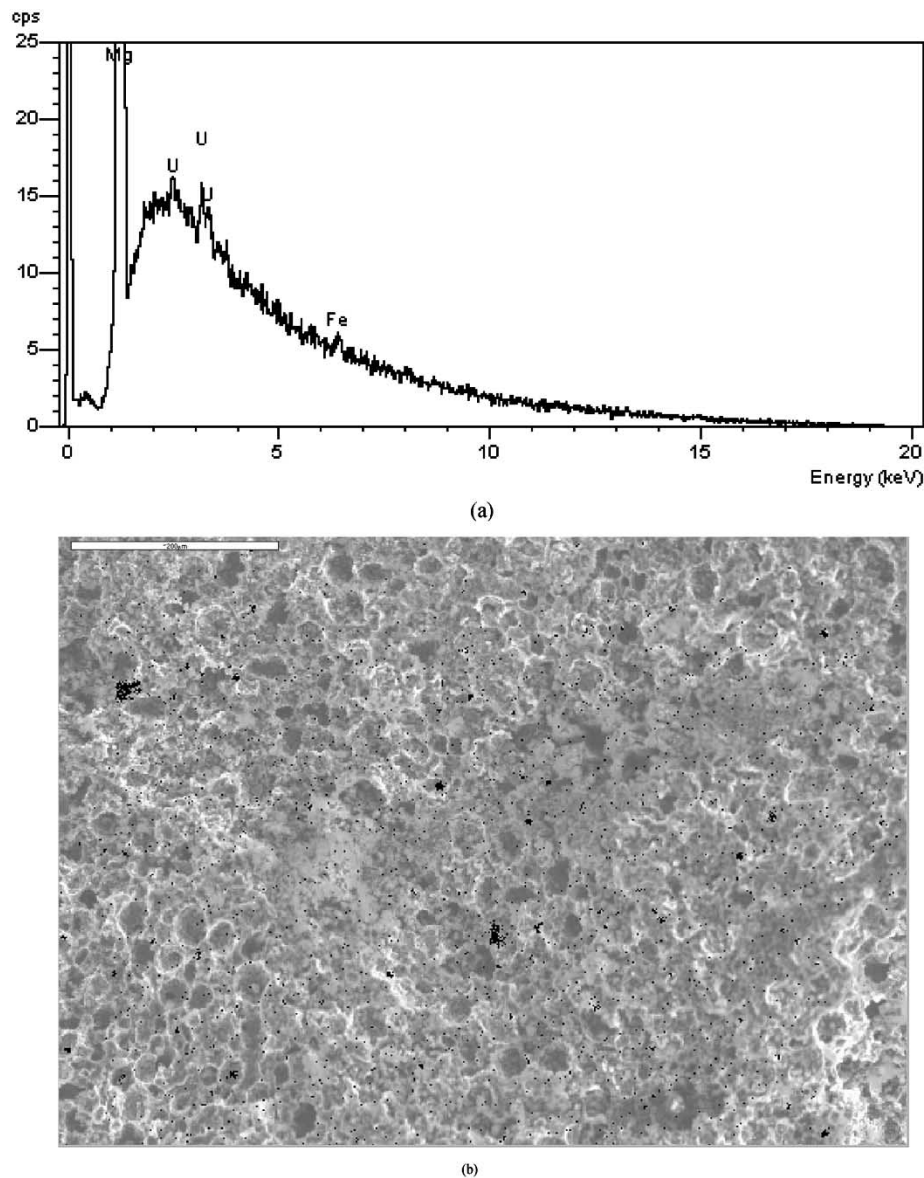


Fig. 2. (a) Averaged EDS spectrum of sample E doped with 1 wt% uranium. (b) EDS mapping of a cross-sectional area of sample E showing the distribution of the  $\text{UO}_2$  particles in black.

and  $b$  are the dimensions of the sample perpendicular to the applied field,  $a < b$ .  $T_c$  was determined from the ac susceptibility measurements.

Small block samples, in doped and undoped pairs, were double encapsulated in titanium capsules and irradiated by neutrons in HIFAR, a 10-MW DIDO class research reactor operated by ANSTO at Lucas Heights, Australia. The irradiation rig was located within the graphite region of the reactor, and thus the neutron energy spectrum was highly thermalized with only a minimal fast and epithermal neutron component. The rig was rated with a nominal neutron flux of  $\sim 1 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ . Four different fluences were used:  $2 \times 10^{16} \text{ cm}^{-2}$  (A),  $5 \times 10^{15} \text{ cm}^{-2}$  (B,E),  $5 \times 10^{14} \text{ cm}^{-2}$  (C), and  $5 \times 10^{13} \text{ cm}^{-2}$  (D). Following irradiation, the  $J_c$  and flux pinning properties of the irradiated pellets were then compared with the pre-irradiated results. The doped irradiated samples were examined by TEM to detect any fission tracks and the extent of fission determined by gamma ray spectroscopy.

### III. RESULTS AND DISCUSSION

XRD using a Philips 1730 phase XRD diffractometer showed that greater phase purity could be obtained using the longer, hotter sintering to fully react the crystalline boron and the magnesium (see Fig. 1). After 3 h at 900 °C, the sintered undoped sample E material (a) is  $\text{MgB}_2$  with traces of Mg and MgO. The sintered 1wt% U-doped sample E material (b) also consists of  $\text{MgB}_2$  with traces of Mg, MgO, and  $\text{UO}_2$ . Fig. 1(c) shows the undoped sintered material characteristic of samples A-D. Because the sintering was shorter and at a lower temperature the Mg and MgO impurity contents are much higher, due to the much larger amount of remaining unreacted crystalline boron. These short sintering conditions are appropriate when amorphous boron is used to make  $\text{MgB}_2$ . The impurities would be expected to give a smaller superconducting volume, but the intragranular properties affected by the U/n method would not be degraded.

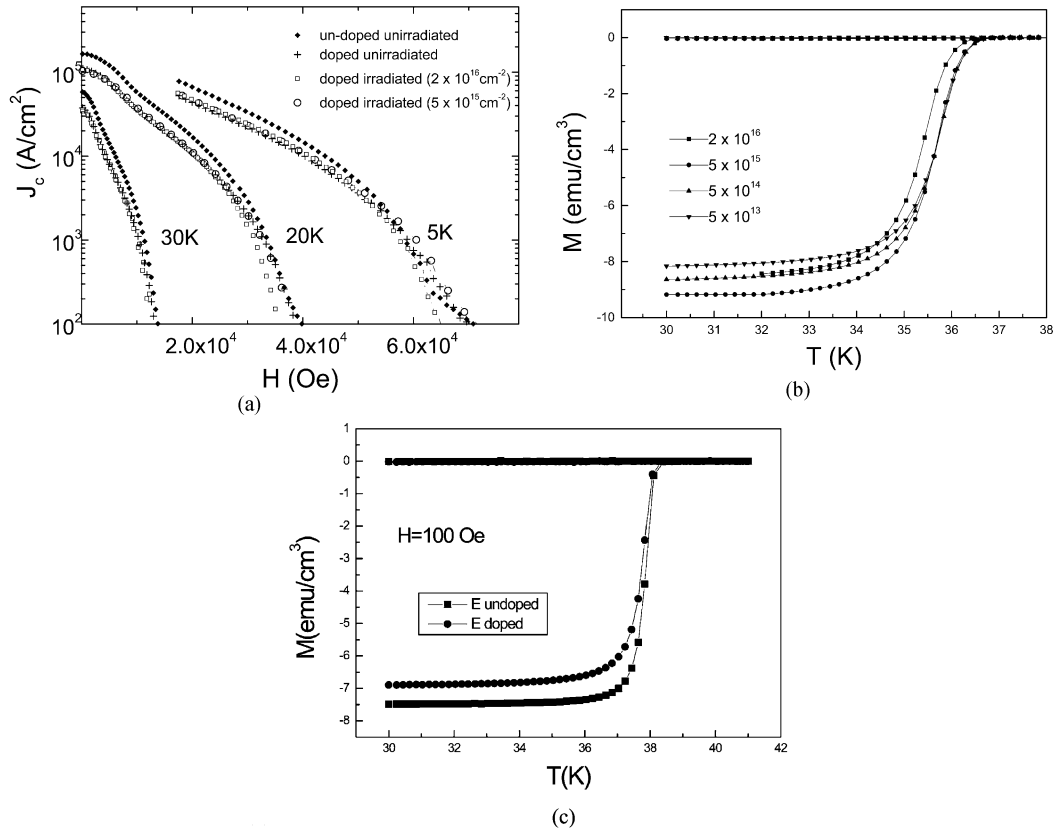


Fig. 3. (a) Critical current derived from PPMS measurements as a function of magnetic field at 5, 20, and 30 K for sample A (undoped and doped with 1 wt% U).  $J_c$  for the doped version of sample A is shown before and after irradiation at  $2 \times 10^{16}$  cm<sup>-2</sup> with thermal neutrons.  $J_c$  for the doped version of sample B (also 1 wt% U) is shown after irradiation with thermal neutrons at  $5 \times 10^{15}$  cm<sup>-2</sup>. Low field 5-K data is not shown as it is affected by flux jumping. (b) Magnetization under ZFC and FC conditions as a function of temperature at a field of 100 Oe as determined by MPMS measurements. All the samples shown were doped to 1 wt% U and irradiated at the fluences/cm<sup>2</sup> that are shown in the legend. (c) MPMS measurements under the same conditions for the unirradiated high purity sample E, both undoped and doped to 1 wt% U.

Samples were characterized using a Cambridge Leica scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS). The results showed MgB<sub>2</sub> grains in a porous structure. The UO<sub>2</sub> particles remained separate, and the larger agglomerates could be distinguished at the grain boundaries. EDS analysis revealed the presence of large amounts of Mg and small amounts of U and Fe, the source of which was most likely the iron tubing containing the samples during sintering. The boron could not be detected with the experimental setup. Fig. 2(a) shows an EDS spectrum of the doped version of sample E. Fig. 2(b) shows an EDS map that reveals a reasonably even distribution of UO<sub>2</sub> particles among the MgB<sub>2</sub> grains.

The ac susceptibility of the samples was measured at temperatures from 5 to 45 K and the magnetization from 5 to 30 K using a physical properties measurement system.<sup>1</sup>  $T_c$  was determined from the ac susceptibility and was not found to be significantly affected by irradiation and doping.  $T_c$  was measured to be 38.5 K with a sharp transition for the higher quality sample E, regardless of doping and approximately 37.2 K for the other samples. The discrepancy is probably because samples A-D have a greater concentration of nonmagnetic impurities, which are known to depress  $T_c$  in MgB<sub>2</sub> [32]. Fig. 3(a) shows the derived values of the critical current at 5, 20, and 30 K as a function of magnetic field for three versions of sample A [ 1) undoped and

unirradiated; 2) doped with 1 wt% U, but not irradiated; and 3) the same piece of doped sample A after irradiation with thermal neutrons at  $2 \times 10^{16}$  cm<sup>-2</sup> and for sample B (doped with 1% U, then irradiated with thermal neutrons at  $5 \times 10^{15}$  cm<sup>-2</sup>).  $J_c$  values for sample E were similar, with a slightly lower  $H_{irr}$ , probably because the lower density cancelled out any benefits due to the higher purity. It was impossible to accurately determine  $J_c$  at 5 K for very low fields because of the presence of thermal flux jumping [33].  $J_c$  performance is slightly worse in the doped samples except at high magnetic fields, probably due to the presence of the additional impurities, but there are no significant differences between the irradiated and unirradiated samples, in contrast to what has been demonstrated for HTS superconductors. The slightly worse performance of the high fluence irradiated sample A at high fields may be due to excessive radiation-induced defects from the higher neutron fluence. Fig. 3(b) shows the ZFC-FC results at 100 Oe as a function of temperature for the doped irradiated versions of samples A-D. There are no significant differences with fluence. Fig. 3(c) shows the preirradiation ZFC-FC results for the doped and undoped versions of sample E. The higher  $T_c$  is attributed to the greater phase purity.

TEM was performed on two specimens (A and B) doped to 1 wt% U and irradiated to neutron fluences of  $2 \times 10^{16}$  cm<sup>-2</sup> and  $5 \times 10^{15}$  cm<sup>-2</sup> respectively. For both materials the microstructure was similar, that is, there were grains of MgB<sub>2</sub>

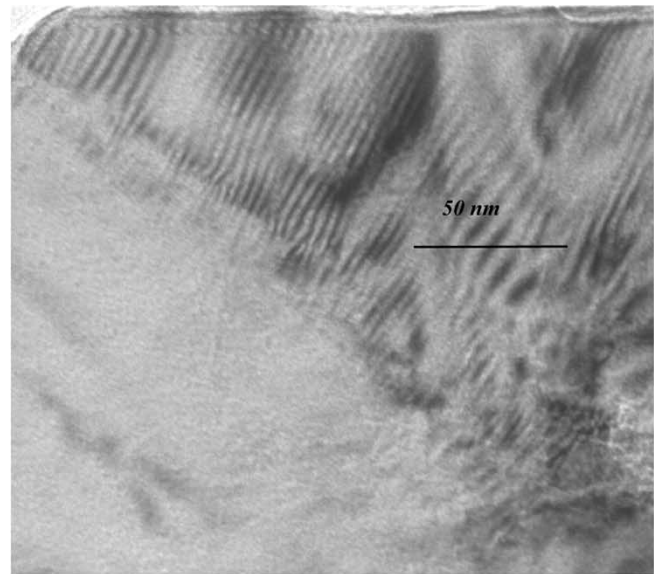
<sup>1</sup>Quantum Design, San Diego, CA.

several microns in diameter in which a number of dislocations and other crystalline defects were evident, but no fission tracks. Many such grains were examined under different imaging conditions, and Fig. 4(a) shows a typical example. This observation is in contrast to TEM studies of U-doped Bi-based HTS specimens exposed to similar levels of irradiation, in which the fission tracks appear as straight randomly oriented black lines due to the greater electron scattering from the amorphous material they contain. These fission tracks are several microns in length and clearly visible in TEM even at modest magnifications. This is illustrated in Fig. 4(b), which shows a TEM image of the core of an irradiated uranium-doped Bi-2223/Ag tape.

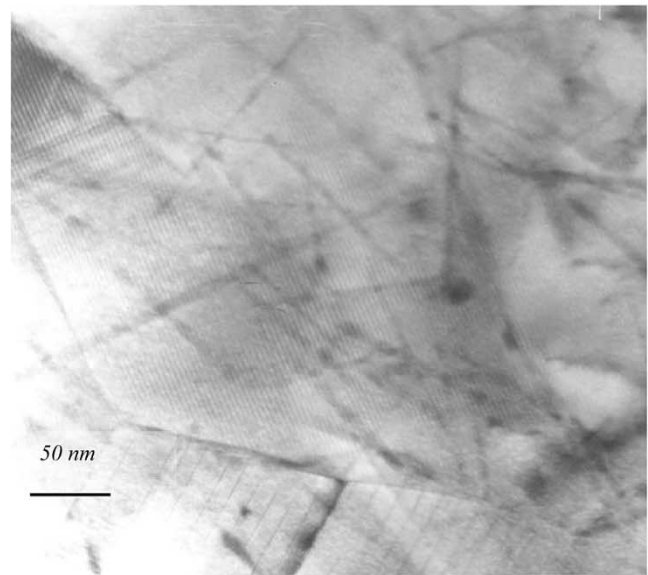
A quantitative analysis of  $^{235}\text{U}$  fissions in the present  $\text{MgB}_2$  samples was obtained from the fission product yield as measured by the gamma-ray spectrometric method [34]. Measurements were limited to the doped irradiated versions of samples C and D. A period of 92 days had expired since irradiation, which meant that most of the short-lived radioisotopes had decayed. This simplified the spectrum. To facilitate measurements, samples were placed 10 cm from the face of a large volume coaxial high purity germanium (HPGe) detector. The gamma ray spectrum for sample C is shown in Fig. 5. The spectral features were consistent with a 96 day decayed  $^{235}\text{U}$  fission product spectrum [35]. The actual fission yield (see Table I) was determined from the measured  $^{137}\text{Cs}$  activity and the cumulative fission yield data for  $^{137}\text{Cs}$  from  $^{235}\text{U}$ . The cumulative yield includes the total number of  $^{137}\text{Cs}$  atoms produced directly as a result of fission in addition to  $^{137}\text{Cs}$  atoms produced via radioactive decay of the  $^{137}\text{Cs}$  precursors, namely  $^{137}\text{I}$  and  $^{137}\text{Xe}$ , which both have half-lives of less than 5 min. By comparison, the half-life of  $^{137}\text{Cs}$  is 30.17 years. The  $^{137}\text{Cs}$  activity was determined from the peak area of the 662-keV emission and the known detector efficiency at this energy.

The fission yield can also be estimated from knowledge of the sample  $^{235}\text{U}$  content, the cross section for fission of  $^{235}\text{U}$ , and the neutron fluence. The estimated fission yield along with the results from the gamma-ray spectrometric method is given in Table I. A good correlation between estimated and measured fission yield was obtained. This shows that nuclear fission of  $^{235}\text{U}$  did occur as expected. In HTS, the same fission would produce columnar defects [Fig. 4(b)], resulting in strong pinning improvement. However, not a single fission track was observed for  $\text{MgB}_2$  [Fig. 4(a)], and the field dependence of  $J_c$  was virtually unchanged after the irradiation [Fig. 3(a)].

In our experiments, heavy ions of about 80 MeV are created by the nuclear fission and transfer their energy to the  $\text{MgB}_2$ . The occurrence or otherwise of the fission tracks can be described by the thermal spike model [36]–[38]. Because of the transfer of energy from the heavy ions to the electrons and subsequently to the crystal lattice on a longer time scale, a sudden localized increase in temperature occurs along the ion path through the crystal. For values of the electronic stopping power  $S_e$  [39] higher than the threshold value  $S_{e0}$  [40], which is dependent on the target material and fission products, the crystal lattice melts in a very localized volume along the ion track. Because the diameter of the molten volume is very small (of the order of a nanometer), this heat is diffused through the crystal lattice quickly enough to freeze the molten volume without allowing recrystallization.



(a)



(b)

Fig. 4. (a) Typical TEM image of an  $\text{MgB}_2$  grain from sample B (doped to 1 wt% U and irradiated at  $5 \times 10^{15} \text{ cm}^{-2}$  with thermal neutrons). This grain, like all the others examined, shows no evidence of columnar defects that might be due to fission tracks. (b) TEM image of the core of a U-doped irradiated Bi-2223/Ag tape showing fission tracks.

Consequently, a track of amorphous material is formed in the crystal, which is not superconducting and is expected to be a strong pinning center. In our experiments, there was no evidence for the formation of fission tracks. The reason for this may be that the  $S_e$  values were too low for the array of fission products interacting with  $\text{MgB}_2$ , or that the heat conductivity required for freezing the molten volume is too low in  $\text{MgB}_2$ , or that the energy transfer from the ions to the crystal electrons and the crystal lattice does not occur on timescales that would enable melting of the lattice. Thus, many parameters, currently unknown for  $\text{MgB}_2$ , are required to predict the presence or absence of amorphous tracks.

It is interesting to compare our results with experiments that employed 2 MeV proton irradiation [14] in order to improve

TABLE I  
COMPARISON OF  $^{235}\text{U}$  FISSION YIELD ESTIMATED PRIOR TO IRRADIATION AND MEASURED AFTER IRRADIATION USING THE GAMMA-RAY SPECTROMETRIC TECHNIQUE

Sample	$^{235}\text{U}$ fission yield (Calculated)	$^{235}\text{U}$ fission yield (Gamma spectrometric method)
C	$1.93 \times 10^{11}$	$1.06 \times 10^{11}$
D	$2.31 \times 10^{10}$	$3.04 \times 10^{10}$

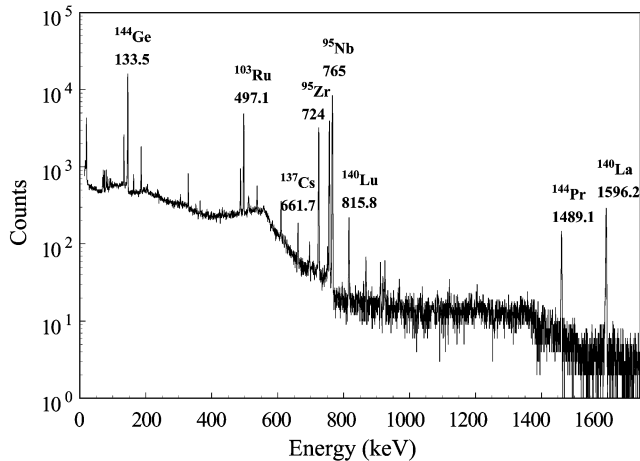


Fig. 5. Gamma ray spectrum of sample C (doped to 1 wt% U and irradiated to a fluence of  $5 \times 10^{15} \text{ cm}^{-2}$ ) 92 days after neutron irradiation.

vortex pinning in  $\text{MgB}_2$ . There, an improvement in the field dependence of  $J_c$  and the irreversibility field was observed. It is likely that the 2 MeV protons used in [14] were producing point defects in  $\text{MgB}_2$ , which are much weaker pinning centers than columnar defects. However, due to the very large density of defects introduced, an observable increase of  $J_c$  was obtained. Point defects are also likely to be responsible for the improvements reported in [15] due to the  $^{10}\text{B}(n, \alpha)^7\text{Li}$  nuclear reaction, rather than any ion tracks.

The relevance of this paper to research in superconductivity is that it shows that uranium fission fragments probably cannot be used for the introduction of columnar defects in  $\text{MgB}_2$ . This research also identifies the need to obtain the values of the parameters affecting the formation of columnar defects in  $\text{MgB}_2$ , if some other method for the introduction of columnar defects is to be devised. It also shows that previously published results on the introduction of pinning centers with high-energy particles have to be taken with caution, because it is highly questionable if columnar defects were introduced in such a way.

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#### REFERENCES

- [1] J. Akimitsu, *2002 Symp. Transition Metal Oxides*, Sendai, Japan, Jan. 10, 2001.
- [2] J. Nagamatsu, J. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, "Superconductivity at 39 K in magnesium diboride," *Nature*, vol. 410, p. 63, 2001.
- [3] C. Buzea and T. Yamashita, "Review of the superconducting properties of  $\text{MgB}_2$ ," *Supercond. Sci. Technol.*, vol. 14, p. R115, 2001.
- [4] C. B. Eom, M. K. Lee, J. H. Choi, L. J. Belenky, X. Song, L. D. Cooley, M. T. Naus, S. Patnaik, J. Jiang, M. Rikel, A. Polyanskii, A. Gurevich, X. Y. Cai, S. D. Bu, S. E. Babcock, E. E. Hellstrom, D. C. Larbalestier, N. Rogado, K. A. Regan, M. A. Hayward, T. He, J. S. Slusky, K. Inamaru, M. K. Haas, and R. J. Cava, "High critical current density and enhanced irreversibility field in superconducting  $\text{MgB}_2$  thin films," *Nature*, vol. 411, p. 558, 2001.
- [5] M. E. McHenry and R. A. Sutton, "Flux pinning and dissipation in high temperature oxide superconductors," *Progress Mater. Sci.*, vol. 38, p. 159, 1994.
- [6] K. H. P. Kim, W. N. Kang, M. S. Kim, C. U. Jung, H. J. Kim, E. M. Choi, M. S. Park, and S. I. Lee, "Negligible effect of grain boundaries on the supercurrent density in polycrystalline  $\text{MgB}_2$ ," *Phys. C*, vol. 370, p. 13, 2002.
- [7] Y. Bugoslavsky, G. K. Perkins, X. Qi, L. F. Cohen, and A. D. Caplin, "Vortex dynamics in superconducting  $\text{MgB}_2$  and prospects for applications," *Nature*, vol. 410, p. 563, 2001.
- [8] H. H. Wen, S. L. Li, Z. W. Zhao, H. Jin, Y. M. Ni, W. N. Kang, H. J. Kim, E. M. Choi, and S. I. Lee, "Magnetic relaxation and critical current density of  $\text{MgB}_2$  thin films," *Phys. Rev. B*, vol. 64, p. 134 505, 2001.
- [9] H. H. Wen, S. L. Li, Z. W. Zhao, H. Jin, Y. M. Ni, Z. A. Ren, G. C. Che, and Z. X. Zhao, "Flux dynamics and vortex phase diagram of the new superconductor  $\text{MgB}_2$ ," *Phys. C*, vol. 363, p. 170, 2001.
- [10] D. C. Larbalestier, L. D. Cooley, M. O. Rikel, A. A. Polyanskii, J. Jiang, S. Patnaik, X. Y. Cai, D. M. Feldmann, A. Gurevich, A. A. Squitieri, M. T. Naus, C. B. Eom, E. E. Hellstrom, R. J. Cava, K. A. Regan, N. Rogado, M. A. Hayward, T. He, J. S. Slusky, P. Khalifah, K. Inamaru, and M. Haas, "Strongly linked current flow in polycrystalline forms of the superconductor  $\text{MgB}_2$ ," *Nature*, vol. 410, p. 186, 2001.
- [11] D. S. Fisher, M. P. A. Fisher, and D. A. Huse, "Thermal fluctuations, quenched disorder, phase transitions, and transport in type-II superconductors," *Phys. Rev. B*, vol. 43, p. 130, 1991.
- [12] S. X. Dou, S. Soltanian, J. Horvat, X. L. Wang, S. H. Zhou, M. Ionescu, and H. K. Liu, "Enhancement of the critical current density and flux pinning of  $\text{MgB}_2$  superconductor by nanoparticle SiC doping," *Appl. Phys. Lett.*, vol. 81, p. 3419, 2002.
- [13] X. L. Wang, S. H. Zhou, M. J. Qin, P. R. Munroe, S. Soltanian, H. K. Liu, and S. X. Dou, "Significant enhancement of the flux pinning in  $\text{MgB}_2$  superconductor through nano-Si addition," *Phys. C*, vol. 385, p. 461, 2003.
- [14] Y. Bugoslavsky, L. F. Cohen, G. K. Perkins, M. Polichetti, T. J. Tate, R. Gwilliam, and A. D. Caplin, "Enhancement of the high-magnetic field critical current density of superconducting  $\text{MgB}_2$  by proton irradiation," *Nature*, vol. 411, p. 561, 2001.
- [15] E. Babic, B. Miljanic, K. Zadro, I. Kusevic, Z. Marohnic, D. Drobac, X. L. Wang, and S. X. Dou, "Enhancement of flux pinning in neutron irradiated  $\text{MgB}_2$  superconductor," *Fizika A*, vol. 10, p. 87, 2001.
- [16] M. Eisterer, M. Zehetmayer, S. Tonies, and H. W. Weber, "Neutron irradiation of  $\text{MgB}_2$  bulk superconductor," *Supercond. Sci. Technol.*, vol. 15, p. L9, 2002.
- [17] H. Narayan, S. B. Samanta, A. Gupta, A. V. Narlikar, R. Kishore, K. N. Sood, D. Kanjilal, T. Muranaka, and J. Akimitsu, "SEM, STM/STS and heavy ion irradiation studies on magnesium diboride superconductor," *Phys. C*, vol. 377, p. 1, 2002.
- [18] .
- [19] R. Weinstein, Y. Ren, J. Liu, I. Chen, R. Sawh, C. Foster, and C. Obot, *Proc. Int. Symp. Superconductivity, Hiroshima, 1993*, Springer, Berlin, Germany, 1993, p. 285.
- [20] G. W. Schulz, C. Klein, H. W. Weber, S. Moss, R. Zeng, S. X. Dou, R. Sawh, Y. Ren, and R. Weinstein, "Enhancement of transport critical current densities in  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  tapes by fission tracks," *Appl. Phys. Lett.*, vol. 73, p. 3935, 1998.
- [21] S. Tönies, H. W. Weber, Y. C. Guo, S. X. Dou, R. Sawh, and R. Weinstein, "On the current transport limitations in Bi-based high temperature superconducting tapes," *Appl. Phys. Lett.*, vol. 78, p. 3851, 2001.

- [22] I. Matsubara, H. Tanigawa, T. Ogura, H. Yamashita, M. Kinoshita, and T. Kawai, "Uppercritical field and anisotropy of the high- $T_c$   $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  phase," *Phys. Rev. B*, vol. 45, p. 7414, 1992.
- [23] S. X. Dou, Y. C. Guo, D. Marinaro, J. W. Boldeman, J. Horvat, P. Yao, R. Weinstein, A. Gandini, R. Sawh, and Y. Ren, "The effects of uranium doping and thermal neutron irradiation on the pinning properties of Ag/Bi-2223 tapes," *IEEE Trans. Appl. Supercond.*, vol. 11, p. 3896, 2001.
- [24] M. Eisterer, S. Tönies, H. W. Weber, R. Weinstein, R. Sawh, and Y. Ren, "High critical currents due to fission tracks in  $\text{Yba}_2\text{Cu}_3\text{O}_{7-\delta}$ ," *Phys. C*, vol. 341–348, p. 1439, 2000.
- [25] T. K. Worthington, W. J. Gallagher, and T. R. Dinger, "Anisotropic nature of high-temperature superconductivity in single-crystal  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ ," *Phys. Rev. Lett.*, vol. 59, p. 1160, 1987.
- [26] J. R. Clem, "Two-dimensional vortices in a stack of thin superconducting films—A model for high-temperature superconducting multilayers," *Phys. Rev. B.*, vol. 43, p. 7837, 1991.
- [27] —, "Phenomenological theory of magnetic structure in the high-temperature superconductors," *Phys. C*, vol. 162–164, p. 1137, 1989.
- [28]
- [29] M. B. Maple, B. J. Taylor, N. A. Frederick, S. Li, V. F. Nesterenko, S. S. Indrakanti, and M. P. Maley, "Critical scaling and flux dynamics in bulk  $\text{MgB}_2$  and high-purity  $\text{Yba}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals," *Phys. C*, vol. 382, p. 132, 2002.
- [30] S. K. Gupta, S. Sen, A. Singh, D. K. Aswal, J. V. Yakhmi, E. M. Choi, H. J. Kim, K. H. P. Kim, S. Choi, H. S. Lee, W. N. Kang, and S. I. Lee, "I-V characteristic measurements to study the nature of the vortex state and dissipation in  $\text{MgB}_2$  thin films," *Phys. Rev. B*, vol. 6610, p. 4525, 2002.
- [31] N. Takezawa and K. Fukushima, "Optimal size of an insulating inclusion acting as a pinning center for magnetic flux in superconductors: Calculation of pinning force," *Phys. C*, vol. 290, p. 31, 1997.
- [32] "Eur. Phys. J. B.," vol. 30, 2002.
- [33] S. X. Dou, X. L. Wang, J. Horvat, D. Milliken, A. H. Li, K. Konstantinov, E. W. Collings, M. D. Sumption, and H. K. Liu, "Flux jumping and a bulk-to-granular transition in the magnetization of a compacted and sintered  $\text{MgB}_2$  superconductor," *Phys. C*, vol. 361, p. 79, 2001.
- [34] "Compilation and evaluation of fission yield nuclear data," IAEA, IAEA-TECDOC-1168, 2000.
- [35] R. L. Heath, "Gamma-Ray Spectrum Catalog," ANCR-1000-2.
- [36] G. Szenes, "General feature of latent track formation in magnetic insulators irradiated with swift, heavy ions," *Phys. Rev. B*, vol. 51, p. 8026, 1995.
- [37] F. Seitz and J. F. Koehler, *Solid State Physics: Advances in Research and Applications*, F. Seitz and D. Turnbull, Eds. New York: Academic, 1995, vol. 2, p. 305.
- [38] M. Toulemonde, C. Dufour, and E. Paumier, "Transient thermal process after a high-energy heavy-ion irradiation of amorphous metals and semiconductors," *Phys. Rev. B*, vol. 46, p. 14 362, 1992.
- [39] Y. Zhu, Z. X. Cai, R. C. Budhani, M. Suenaga, and D. O. Welch, "Structures and effects of radiation damage in cuprate superconductors irradiated with several-hundred-MeV heavy ions," *Phys. Rev. B*, vol. 48, p. 6436, 1993.
- [40] J. Provost, Ch. Simon, M. Hervieu, D. Groult, V. Hardy, F. Studer, and M. Toulemonde, "Swift, heavy ions in insulating and conducting oxides: Tracks and physical properties," *Mater. Res. Soc. Bulletin*, vol. 20, p. 22, 1995.
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