



# Irradiation effects on $\text{MgB}_2$ bulk samples and formation of columnar defects in high- $T_c$ superconductor

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## Abstract

Irradiation effects on  $\text{MgB}_2$  were studied to investigate the pinning property of this new material. It is pointed out that flux pinning at grain boundaries is important for  $\text{MgB}_2$  bulk samples. We confirmed this with electron irradiation. Degradation of inter-grain coupling by electron irradiation resulted in degradation of pinning properties. Heavy ion irradiation was also accomplished on  $\text{MgB}_2$ . Columnar defects introduced by the irradiation improve pinning in higher field regions. Concerning the formation of columnar defects in high- $T_c$  superconductors, we conducted a systematic analysis of  $\text{Bi2212}$  single crystal with different irradiation ions. Applying the time dependent line source model to our results, only a third of the electronic losses contributed to the formation of columnar defects.

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## 1. Introduction

A new superconducting material  $\text{MgB}_2$  has the highest transition temperature of conventional inter-metallic superconductors [1]. The working temperature is almost 40 K, and helium-free ap-

plications using refrigerators are expected. Furthermore, this material consists of light atoms and has an advantage for industrial usage. This new material, however, shows a rapid decrease of superconductivity in magnetic fields. Improvements of the pinning properties are necessary for practical applications. In this work, we investigated irradiation effects on this material. Columnar defects are well known as effective pinning centers, but the mechanism of their formation is not clear. We also investigated the formation mechanism of the columnar defects in high- $T_c$  superconductors.

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## 2. Irradiation on MgB<sub>2</sub>

Synthesis of MgB<sub>2</sub> was accomplished by a solid state reaction. High purity powders of Mg and B were used for the synthesis. They were mixed well and ground, then pressed into a small rod (8 mm in diameter and 80 mm in length) for the sintering process. The rod was placed into a stainless tube, and the tube was put into a tube furnace. The sample was sintered at 1193 K for 2 h. Throughout the sintering process, high purity argon gas was flowed in the furnace to avoid unexpected reactions, such as MgO or BN. After the sintering, the sample was cut into small pieces. The magnetic properties were measured by a commercial SQUID magnetometer (MPMS, Quantum Design). One and the same sample was used for individual irradiations to compare the differences of pinning properties between pre- and post-irradiation conditions.

Some of the earliest studies on the pinning properties of MgB<sub>2</sub> indicate that grain boundaries are important for improvement of the critical current density ( $J_c$ ) [2,3]. Electron irradiation was used to confirm this. Electron irradiation of sintered materials usually affects the grain boundaries. Thus, the critical current density should decrease after the irradiation because of degradation among the boundaries. The irradiation at 2.5 MV accelerating voltage was achieved in the Japan Atomic Energy Research Institute (JAERI), Takasaki establishment up to the irradiation fluence of  $5 \times 10^{17}$  e/cm<sup>2</sup>. The inset of Fig. 1 shows the change in  $J_c$ . No enhancement on  $J_c$  can be observed with the irradiation. For all temperatures, little difference was observed in low fields. On the contrary, the critical current densities decrease at higher fields after the irradiation. Thus, the irradiation degrades the bulk pinning properties of this material. The difference in susceptibilities of the shielding currents (after zero-field cooling) between before and after the irradiation is shown in Fig. 1. It is found that the pinning properties decrease as the shielding currents decrease. This suggests that the decrease of surface pinning due to the degradation of superconducting coupling among the grains occurs by the electron irradiation and vortices can penetrate easily into the interior of the sample.

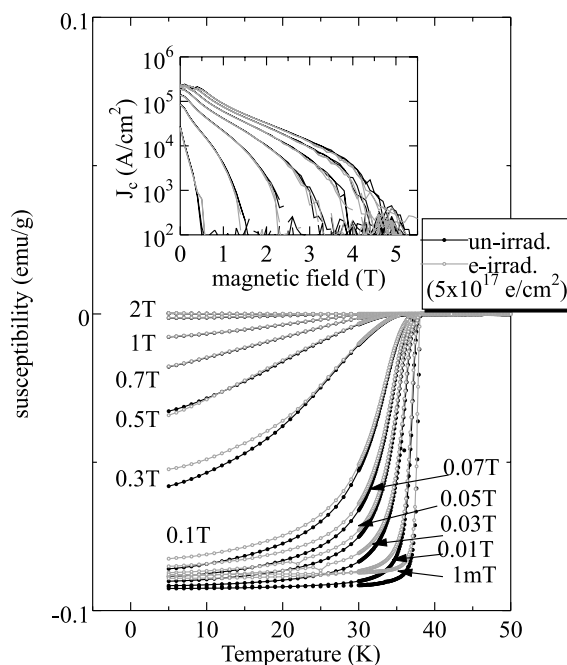


Fig. 1. Susceptibility comparison between before and after electron irradiation. Applied fields are 2, 1, 0.7, 0.5, 0.3, 0.1, 0.07, 0.05, 0.03, 0.01 T and 1 mT, respectively. Inset (upper part of figure): Critical current densities before and after electron irradiation. Measuring temperatures (from upper right to lower left) are 5, 10, 15, 20, 25, 30 and 35 K, respectively.

Columnar defects introduced by heavy ion irradiation are well known as strong pinning centers to greatly improve pinning properties of high- $T_c$  superconductors [4]. It is worth confirming the effectiveness of the columnar defects on pinning for the new material. Heavy ion irradiation was accomplished at the ring cyclotron in RIKEN with 3.54 GeV Xe ions. The irradiation dose was  $1 \times 10^{11}$  ions/cm<sup>2</sup> corresponding to the matching field  $B_\phi = 2$  T. Fig. 2 shows the comparison of the critical current density between before and after irradiation. No enhancement of  $J_c$  in the lower field region can be observed in any temperature range, however, the irreversibility field shifts to the higher field region. For high- $T_c$  superconductors, columnar defects improve both the magnitude of the critical current density and the irreversibility field. For MgB<sub>2</sub>, they improve only the latter. Columnar defects act as good pinning centers. Therefore, it is possible to improve the pinning

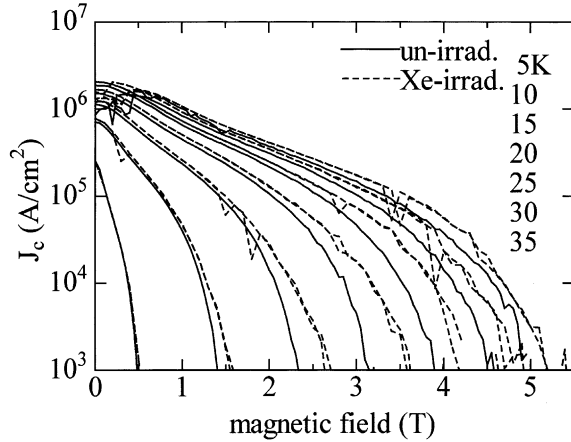


Fig. 2. Critical current densities before and after heavy ion irradiation. Measuring temperatures (from upper right to lower left) are 5, 10, 15, 20, 25, 30 and 35 K, respectively.

properties of MgB<sub>2</sub> by introducing appropriate pinning centers.

### 3. Formation mechanism of columnar defects in high- $T_c$ superconductors

As mentioned above, columnar defects are effective for improving the pinning properties. When a bombarding high-energy ion passes through the high- $T_c$  sample, it loses its energy, exciting electrons within a narrow cylindrical region along its trajectory. The excited high-energy electrons then transfer their energies to the lattice. If the transferred energies are large enough, the lattice of the target will melt partially along the ion trajectories and columnar defects will be formed. Thus there are two important parameters in understanding columnar defect formation: one is the energy transfer rate from bombarding ion to the electron system,  $(dE/dx)_e$  and the other is the diameter of the columnar defect,  $R_o$ . To investigate the relation between  $(dE/dx)_e$  and  $R_o$ , we irradiated Bi2212 single crystal ( $T_c = 90$  K) with different energy Au ions (from 120 to 300 MeV). The irradiations were accomplished at the tandem accelerator in the JAERI Tokai establishment. Samples were irradiated up to a dose of  $6.3 \times 10^{10}$  ions/cm<sup>2</sup> corresponding to the matching field  $B_\phi = 1.3$  T (the

irradiating direction was parallel to the  $c$  axis). The diameters of the columnar defects were observed by transmission electron microscopy (TEM). The energy transfer rate of the individual ion energy was calculated by the TRIM code [5]. The values obtained for each  $(dE/dx)_e$  and  $R_o$  at different ion energies are listed in Table 1. The relation between  $(dE/dx)_e$  and  $R_o$  was evaluated with a modified thermal spike model (time dependent line source model, TDLS) developed by Izui [6]. This model was successfully applied to the interpretation of the formation mechanism of columnar defects in semiconductors. According to Izui's model, temperature  $T(r, t)$  at a point with normal distance  $r$  from a line heat source at the time  $t$  is expressed as [7,8]

$$T(r, t) = \frac{1}{4\pi K} \int_0^t \frac{Q(t')}{t-t'} \exp \left\{ -\frac{r^2}{4D(t-t')} \right\} dt', \quad (1)$$

where  $K$  and  $D$  are the thermal conductivity and thermal diffusion coefficients, respectively. Here, time-dependent function  $Q(t)$  is the thermal energy release rate transferring from excited electrons to the lattice via electron–phonon interactions. The equation that an electron transfers its energy to the lattice is given by [9]

$$-\frac{d\varepsilon}{dt} = \left( \frac{4}{9\pi} \right) \left( \frac{C^2 m}{\hbar N M} \right) k^3, \quad (2)$$

where  $k$  is the wave number of the electron,  $N$  is the density of atoms in the lattice,  $m$  is the electron mass,  $M$  is the atomic mass, and  $C$  is a coupling parameter characterizing the electron–phonon interaction. Thus  $Q(t)$  is given by  $Q(t) = -n(d\varepsilon/dt)$ , where  $n$  is the number of contributing electrons

Table 1  
Irradiation parameters obtained from our experiments

Ion	Energy (MeV)	$S_e$ (keV/nm)	$R_o$ (nm)
Au <sup>+</sup>	120	19.0	9.0
Au <sup>+</sup>	180	23.0	10.5
Au <sup>+</sup>	240	26.8	12.5
Au <sup>+</sup>	300	29.0	16.0

The deposited energy due to electron excitation  $S_e$  is calculated by the TRIM code. The diameters of the columnar defects are obtained from our TEM observations.

within unit length along the line source. Using the relation  $\varepsilon = (\hbar k)^2/2m$ ,  $Q(t)$  is expressed explicitly as a function of  $t$

$$Q(t) = -n \frac{d\varepsilon}{dt} = nA \left( \frac{A}{2}t + \varepsilon_0^{-1/2} \right)^{-3},$$

$$A = \frac{8\sqrt{2}m^{5/2}C^2}{9\pi\hbar^4NM},$$

where  $\varepsilon_0$  is the initial energy which an electron receives from an incident ion.

Applying this model to our experimental results, it was found that the deposited electron energy ( $S_e$ ) calculated by the TRIM code was too large. Thus we tried to find the effective deposited energy for columnar defect formation,  $w$ , as a parameter. We assumed that columnar defect formation occurs under the condition that the lattice temperature at  $r$  (the edge of the columnar defect) is heated up above 1000 °C (melting point)

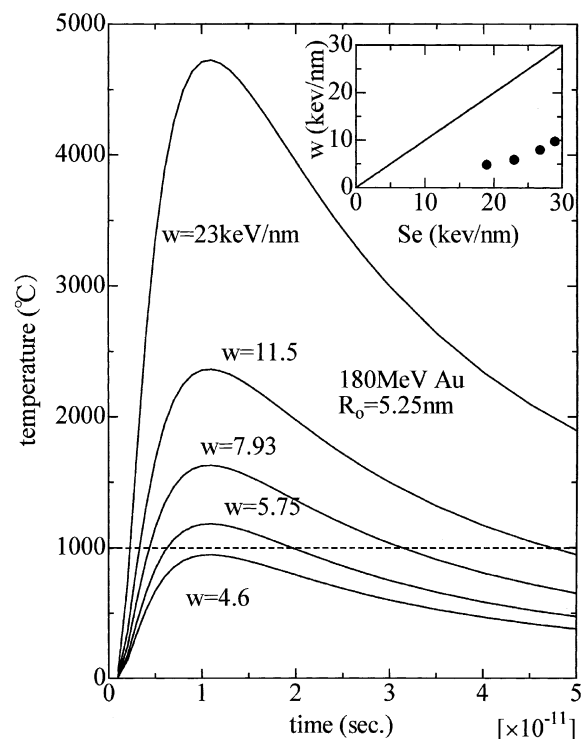


Fig. 3. Calculation of temperature change at the edge of the columnar defect based on the TDLS model. Effective deposited energy for columnar formation  $w$  is a parameter. Inset: the relation between  $S_e$  and  $w$ .

during  $5 \times 10^{-12}$  s [10]. We choose the minimum value satisfying the above conditions as  $w$ . The result is shown in Fig. 3 for the 180 MeV Au ion irradiation case. The effective deposited energy is 5.75 keV/nm, only a fourth of  $S_e$ . For all cases of our experiments,  $w$  is in the range from a third to a fourth of  $S_e$  (see inset of Fig. 3). Only a small part of  $S_e$  is used for columnar defect formation and most of  $S_e$  vanishes. This is caused by the large anisotropy of high- $T_c$  superconductors. Deposited energies in the  $\text{CuO}_2$  plane diffuse immediately and only that deposited along the  $c$  axis contributes to columnar defect formation.

#### 4. Conclusion

Irradiations on  $\text{MgB}_2$  bulk samples have been accomplished. Electron irradiation affects the inter-grain coupling. Heavy ion irradiation introduces columnar defects in  $\text{MgB}_2$  and the irreversibility field is improved. Columnar defect formation in  $\text{Bi2212}$  is considered using the TDLS model. The effective energy deposition for columnar defect formation is much smaller than  $S_e$ .

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#### References

- [1] J. Nagamatsu et al., Nature 410 (2001) 63.
- [2] Y. Takano et al., App. Phys. Lett. 78 (2001) 2914.
- [3] Y. Bugoslavsky et al., Nature 411 (2001) 561.
- [4] J.R. Thompson et al., App. Phys. Lett. 60 (1992) 2306.
- [5] J.F. Ziegler, Handbook of Stopping Cross Section for Energetic Ions in All Elements, Pergamon, New York, 1980.
- [6] K. Izui, J. Phys. Soc. Jpn. 20 (1965) 915.
- [7] M. Sasase et al., Advances in Superconductivity XII, Springer-Verlag, Tokyo, 2000, p. 314.
- [8] H.S. Carshaw, J.C. Jaeger, Conduction of Heat in Solid, Clarendon Press, Oxford, 1959, p. 261.
- [9] F. Seitz, Phys. Rev. 74 (1949) 1376.
- [10] G.A. Rozgonyi et al., Laser and Electron-Beam Interactions with Solids, North-Holland, New York, 1982, p. 177.