Magnetic instability in irradiated MgB_2 dense samples

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High density magnesium diboride samples were irradiated with low dosages of γ-rays, protons, and electrons. They were investigated by magnetization and thermal studies in order to determine if the irradiation increases the flux pinning and consequently the critical current density Jc. Zero field cooled magnetic susceptibility and specific heat measurements confirm the bulk transition temperature (Tc) with diamagnetic signal at $\sim$38.5 K. Magnetic instabilities were observed in the superconducting hysteresis loop at temperatures between 2 and 23 K in all studied polycrystalline MgB_2 dense samples. The occurrence of flux jumps depended of the type of irradiation and the number of jumps decreases as temperature increases. The critical current density Jc, estimated from the magnetization hysteresis using the Bean's model, is improved for gamma irradiated sample at $H=0$ and $T=2$ K. At low temperature, the Jc decreases and several steep drops in Jc are observed as a function of applied magnetic field. Furthermore, it is observed that the influence of crystalline defects plus local disorder, induced by hot isostatic pressure and irradiation with energetic atoms, increase the Jc but at the same time the magnetothermal instabilities increase in a broad range of temperature. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3008027]

I. INTRODUCTION

MgB_2 is a new and important superconducting material in which many work has been done in order to study its basic and technological properties. In the past years, the study of MgB_2 has changed from basic to applied research in terms of improving the critical current density (J_c) for practical purposes. Therefore, several methods have been developed to increase the Jc of this compound, for instance, the doping with nanoparticles as carbon nanotubes,[1,2] nano-SiC,[3] nano-Fe,[4] and nano-Co_3O_4 (Ref. 5) as well as ion irradiation. These procedures have been frequently used for inducing crystalline disorder and hence pinning centers.[6,7] The mechanism is to create pinning centers optimally distributed in the superconductor to quench the motion of vortices and to inhibit the rapid decline of Jc with increasing the field strength. The research in producing high quality of MgB_2 dense bulk,[8] thin films,[9] and wires[10] with the capability of carrying higher current density is a technological topic in progress. However, factors such as thermal instabilities produce negative effects on the current carrying capabilities.

In magnetization versus applied magnetic field (M-H) experiments have found flux jump behavior that is disadvantageous because it reduces the Jc, limiting the superconducting applications.[11] Magnetization jumps are usually associated with thermomagnetic instability of the flux line (vortices) penetrating the material. In other words, vortex motion is a mechanism of heat dissipation, and the local temperature will rise tending to depinning of the vortex lattice, leading to Joule heating.[12] This process in a sample region produces sudden catastrophic decreases on the magnetization, which can be considered as large macroscopic scale avalanches. This effect has been observed in Pb, Nb, and recently in MgB_2 thin films and bulk superconductors.[6,11,13,14]

Theoretically and experimentally, it was demonstrated that the introduction of disorder can strongly modify the physical MgB_2 properties, mainly the critical current density. Ion irradiation has been frequently used as a procedure for inducing crystalline disorder and vortex pinning in superconducting materials. The energetic ions displace atoms from their equilibrium lattice site, creating vacancies and interstitials atoms. Such defects tend to depress the superconducting order parameter and thereby creating pinning sites. Thus, we expect that the magnetization and specific heat measurements on highly dense MgB_2 bulk would provide additional useful information for understanding which process dominates the flux motion and subsequent stability of the critical current and if the ion irradiations have effects on both processes.

In this work, we used hot isostatic pressed-MgB_2 samples that were irradiated with low fluxes of electrons, protons, and gamma-rays. M versus H at different temperatures and heat capacity measurements were performed and

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the results were compared with the pristine sample. All samples investigated display magnetothermal instabilities, as well defined "flux jumps" up to 23 K. The flux jumps in the magnetization hysteresis loop as well as the corresponding critical current density Jc depend on the type of irradiation and they increase as temperature is decreased. At 10 K and zero magnetic field the Jc has been improved for irradiated samples with respect to pristine sample. Furthermore, the number of flux jumps decreases with temperature and disappears at about 23 K. Based in this and other results we speculate that the flux jump seems to occur as a results of high critical current and very low heat capacity closely connected with very high defect density, which is induced by the preparation sample method (hot isostatic pressed) and the irradiation damage on the sample. These findings should be considered as an inconvenient for potential future applications.

II. EXPERIMENTAL DETAILS

Samples were prepared using hot isostatic pressing method with a pressure of 200 MPa. The technical details about the procedure for produce dense samples are given elsewhere and are omitted here for brevity.[8] This procedure yields high density MgB_2 bulk samples (2.66 ± 0.004 g/cm^3), which appeared to be higher than the theoretical density based on x-ray measurements (2.625 g/cm^3). Three MgB_2 polycrystalline samples were irradiated independently with electrons, protons, and gamma-rays. Electron and proton fluxes were produced with 2 MeV van de Graaf accelerator (high voltage Engineering Corp.) and a 3 MeV Pelletron accelerator 9SDH, respectively. For the case of gamma-rays, the source of irradiation came from an excitation source of ^{60}Co (gamma beam 651 PT Nordion International Inc.). The irradiation conditions were the following dose rate: 25 kGy/min with total dosage of 5000 kGy, 244 MGy as total dosage, and 9.3 kGy/h with a total dosage of 5000 kGy for electron, proton, and gamma irradiation, respectively. It is good to underline that the penetration depth for γ-irradiated samples covers the complete sample and passes through it. For electron irradiation, it depends on the density of the material as well as the energy employed for the irradiation. Henceforth, for the information provided formerly, the electron penetrates about 3 mm into the sample. Moreover, for the proton implantation and for the 3 MeV energy, the calculated penetration was 93 μm. The bulk dense samples were extracted and the magnetization and specific heat was measured using a commercial Quantum Design superconducting quantum interference device magnetometer and Quantum Design physical properties measurement system (1 T), respectively. dc magnetization was measured in applied magnetic field of 10 Oe and the magnetization loops were obtained with applied magnetic fields up to ± 4 T. Furthermore, specific heat was measured down to 2 K in zero applied field.

III. RESULTS AND DISCUSSION

Figure 1 yields the volume diamagnetic susceptibility signal $1/4\pi$ at low temperature in an applied magnetic field of 10 Oe for pristine and irradiated MgB_2 samples. The onset

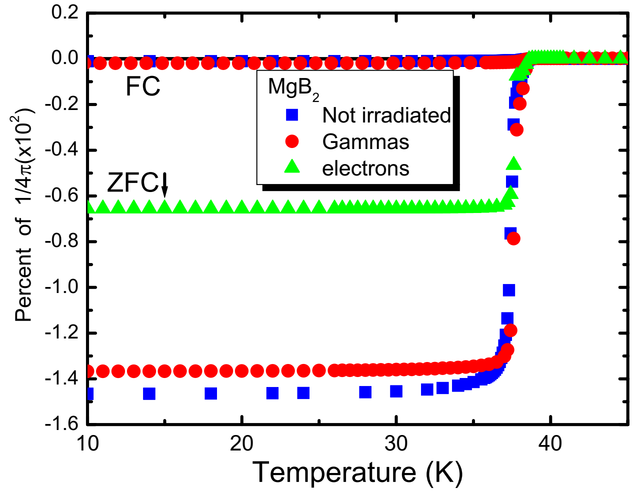

FIG. 1. (Color online) Magnetic susceptibility as a function of temperature expressed in terms of the flux expulsion.

of superconductivity is located at ~38.5 K in the three samples, indicating that the irradiation doses have not any effect on the critical superconducting temperature Tc. At 2 K the diamagnetic susceptibility for the field cooled (FC) measurements is ~2% of $1/4\pi$ and in the zero FC the shielding fraction is ~146% and ~138% for pristine and gammas irradiated samples, respectively. A different situation is observed for the electron irradiated samples where the shielding fraction is reduced to ~56% with respect to the nonirradiated sample. The improvement in the bulk superconductivity for the pristine sample is attributed to the compactly connected MgB_2 grains. If the bulk sample is fully superconducting, the shielding value deviating from 100% is likely due to the demagnetization factor, which is not taken into account. However, the reduction in the shielding fraction in the electron irradiated sample is attributed not to connected grains or grain size but very likely to the enlarged penetration length of the external field in the surface of the sample. This indicates that the doses of electron irradiated samples affect the bulk superconducting but not the critical transition superconducting temperature.

A direct quantitative comparison of the effect of irradiation on the bulk superconducting properties can be evaluated by specific heat measurements. Figure 2(a) shows the total specific heat C_p and in Fig. 2(b) is shown the electronic specific heat (C_e) as a function of temperature in the absence of magnetic field for pristine, proton, and electron irradiated sample. At room temperature, the molar specific heat curves of irradiated samples overlap each other, unlike what occurs for pristine sample. The Cp reaches ~39 J/mol K for the pristine sample that corresponds to ~53% of the Dulong–Petit equipartition value ($C_p \sim 3$ R), which is increased for the irradiated sample to about 70%. This splitting of the Cp from 100 to 300 K is a crude appreciation of the effect of the energetic ions irradiation on the host lattice. However, at low temperature, differences in the electronic specific heat behavior are clearly observed. The curves between 5 and 40 K in Fig. 2(b) show a typical experimental specific heat profile. It was demonstrated that the two gaps structure is responsible for the particularities of the heat capacity observed at low

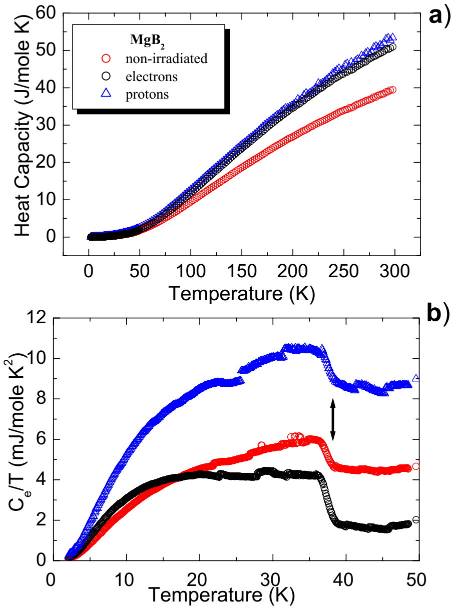

FIG. 2. (Color online) (a) Specific heat for MgB_2 for pristine and irradiated samples from 2 to 300 K. (b) Electronic specific heat in the low temperature region. Arrows indicate the two typical anomalies at about 38.5 and 10 K, which have been ascribed as two superconducting gaps in MgB_2.

temperature and close to Tc. At low temperatures an excess in specific heat profile is showed at about 10 K, which does not follow the conventional single-gap BCS behavior.[15,16] Unfortunately, a quantitative thermodynamical analysis was not possible since the Cp in applied magnetic field data was not possible to obtain; however, qualitative information can be extracted. The transition from the normal to the superconducting state is seen as a discontinuity in the C_p/T at about 38.5 K, which corresponds to the diamagnetic signal discussed above. We observe that only the magnitude of the C_p/T curve is affected by the irradiation and the Tc remains unchanged. For instance, the protons irradiated sample shows a further robust profile of the C_p/T curve in a whole range of temperature being more pronounced around Tc with respect to the electron and pristine sample. A rough estimation of the electronic constant (γ_e) just above of the Tc gives values of 8.44, 1.7, and 4.4 mJ/mol K^2 for proton, electron, and pristine samples. These values are different to that of 2.5–3 mJ/mol K^2 reported for MgB_2.[17,18] Furthermore, the high dispersion in the C_p/T curve around the Tc implies that the irradiation have a strong effect over the larger gap (σ-gap) as well as strong effect in the phonon dispersion, which is inferred by changes in the magnitude and the jump of the $\Delta C_p/T$. Fisher *et al.*[19] showed that the lattice contribution is relatively large around the Tc in MgB_2 and any deviation in the lattice contribution from harmonic-lattice contribution ($C_l = B_3T^3 + B_5T^5$) around Tc should be associated to phonon spectrum, which is known to play an important role in the superconducting properties.

Figures 3(a) and 3(b) show the magnetization as a function of external magnetic field at 2, 10, 15, and 20 K for pristine and irradiated samples. As can be seen in each column of both figures, the corresponding irradiated sample is compared with the nonirradiated sample at the same scale of applied magnetic field and temperature. The irradiation effect is clearly seen in the number of flux jumps and the magnitude of the magnetization in the hysteresis loop cycle at different temperatures measured. Thus, at 2 K, a high number of flux jumps are observed in each quadrant of the hysteresis loop for all samples. The first large jump in the pristine magnetization curve is practically independent of the irradiation and occurs at about 0.8 T; however, this flux jumps are followed by a series of smaller jumps persisting up to 4 T for electron and gamma irradiated samples. It is worth noting that the magnetization instabilities have magnitudes in the order of 500 emu/cm^3 at $H=0$ and $T=2$ K, being higher (~750 emu/cm^3) for electron irradiated samples. In addition, the electron irradiated sample display further magnetic instabilities as the external magnetic field is increased. The flux jumps were observed up to 20 K for all samples as seen in Fig. 3(b) and in the case of electron irradiated sample up to 25 K (not shown). Furthermore, it is observed that the number of flux jumps increases as decreasing temperature shows more magnetic instabilities for gamma and electron irradiated samples. When the temperature is increased at about 10 K, the magnitude of the magnetization (at $H=0$) is higher in all samples reaching ~1750 emu/cm^3 for electron irradiated sample. Again, the first and third quadrant is the more unstable since the flux jumps are present even at 20 K. The magnetic flux trapped plus the increasing of the external magnetic field perturbs the critical state as further flux jumps in the hysteresis magnetization curves.

Figure 4 shows the filed dependence of the nominal critical current Jc for $T=2$, 10, 15, and 20 K. The data were extracted using the Bean model[20] taking into consideration the average dimensions of an irregular parallelepiped shaping in accordance with the formula $J_c(\mathrm{H}) = 20\,\Delta M/(a\text{-}a^2/3b)$, where ΔM (emu cm^{-3}) comes from the magnetization hysteresis loop and the "a" and "b" from the square dimensions of the parallelepiped sample. Although the Bean critical state model could be inapplicable for a flux jump system, we can make an estimation of the nominal Jc in the more stable quadrant for a qualitative comparison. At 2 K, note that all samples show instabilities in critical current capability as applied field is increased. Nevertheless, it is possible to evaluate the applied magnetic field dependence of Jc at several temperatures and to analyze the effect of the irradiation on the Jc. At 10 K and $H=0$ T, the gamma irradiated sample reach values about 2.0×10^6 A/cm^2, which is higher than the Jc value (4.67×10^5 A/cm^2) of the pristine sample. Furthermore, proton and electron irradiation samples show a sharp drop in the Jc in the range of 0–1 T of magnetic field. However, no further instability in the Jc is observed at 15 and 20 K with appreciable and similar improving of the Jc values in proton and electron irradiation with respect to the pristine sample between 0 and 3 T. The Jc values as a function of the temperature at applied field of 0 and 4 T can be seen in Fig. 5. The dense nonirradiated sample shows a critical current in the order of 2.9×10^5 in $H=0$, while the gamma irradiated sample reach to 8.72×10^5 A/cm^2 before the first flux jumps occurs. Also it is observed that the Jc values are higher at 10 K for all studied samples as a result of the decreasing flux jumps number. At 4 T, the Jc decline faster for the proton irradiated sample above of 10 K. Fur-

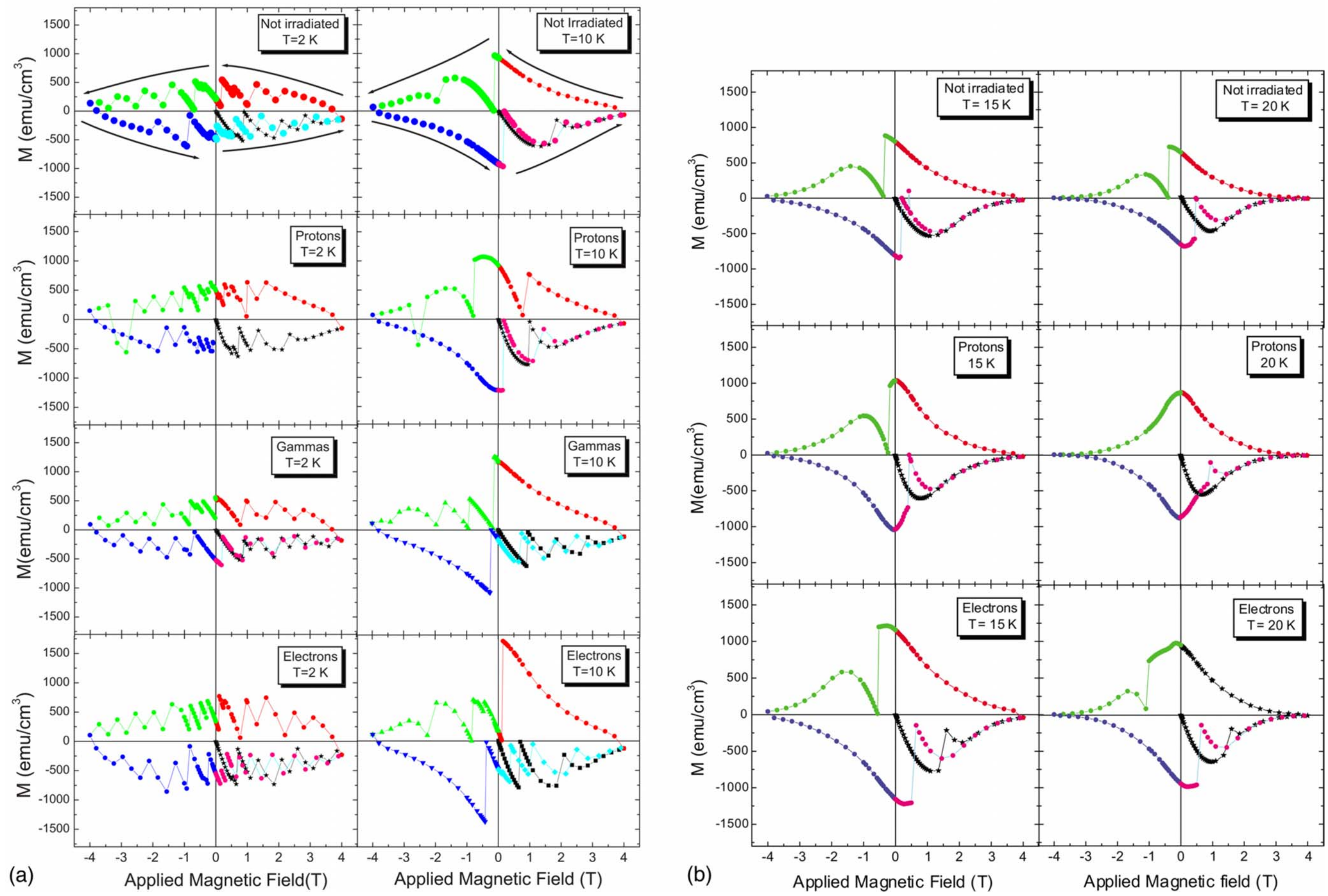

FIG. 3. (Color online) (a) Magnetization vs applied magnetic field at 2 and 10 K for pristine sample as well as proton gammas and electron irradiated samples. The arrows in the top panel indicate the direction of external magnetic field. (b) Magnetization vs applied magnetic field at 15 and 20 K for the same samples. For comparison, the hysteresis loops are in the same scale.

thermore, we can infer that the irradiation damage produced by electrons and protons is similar since the values of Jc show similar behavior as a function of temperature and external magnetic field. In contrast, the gamma irradiation affects strongly the Jc behavior. This result reflects the strong pinning force, or in other words, the notable amount of lattice defects produced by the gamma irradiation when it is compared to proton and electron irradiation effects. This en-

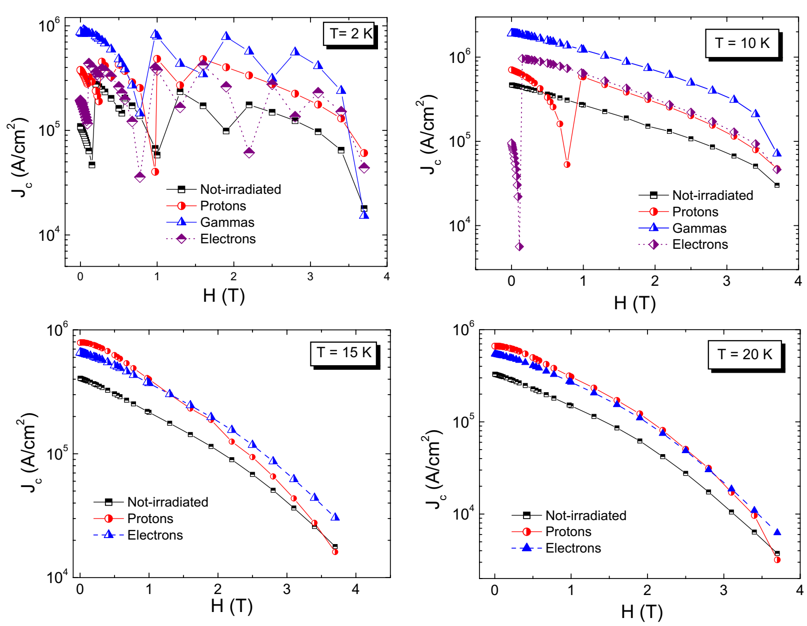

FIG. 4. (Color online) Critical current density Jc as a function of applied magnetic field at 2, 10, 15, and 20 K for pristine MgB_2 sample and samples irradiated with protons, electrons, and gamma-rays.

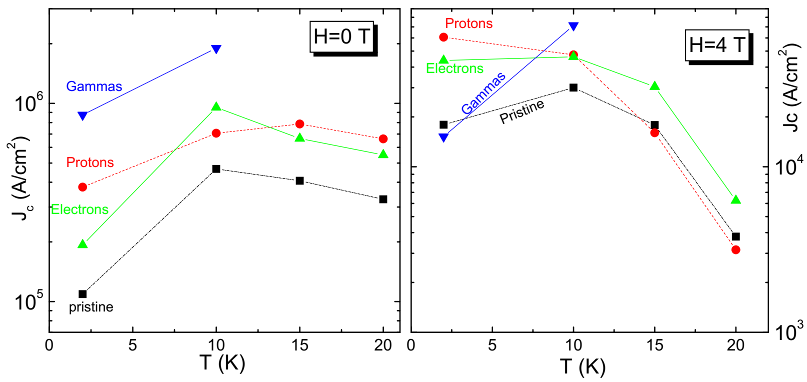

FIG. 5. (Color online) Temperature dependence of the critical current density (Jc vs T) at $H=0$ and 4 T for the pristine and irradiated MgB_2 dense samples (the line is guide for the eyes).

hanced pinning force may be attributed to the energetic characteristic of the irradiation source since gamma irradiation passes through the dense sample, while the damage is superficial with protons and electrons irradiation.

It is important to point out that the flux jumps process is an open problem that deserves more fundamental and experimental investigation. Here, we briefly discuss the close connection between sample bulk processing, epitaxial thin films, and doping nanoparticles with the magnetothermal instabilities. Dou *et al.*[4] and Mudgel *et al.*[21] showed a close connection between high critical current density and flux jumps in nanodoped MgB_2 samples. They assumed that the magnetothermal instabilities occur when effective pinning centers have been induced and consequently extremely high density current resulting in strong coupling between Jc, low specific heat, and magnetothermal instabilities in the form of flux jumps. Recent magneto-optical[22,23] studies on thin films show that avalanches grow gradually from the surface to the inside of the sample in the form of dendritic structures. Qualitatively, these avalanches may correspond to each steep drop in the magnetization in the $M(H)$ loops. On the other hand, a theoretical-experimental approach carried out by our group[24] based on adiabatic critical state model, showed that the flux jumps depend on the ratio of the Jc after the flux jumps and the Jc before the flux jumps. It is found that some amount of applied magnetic field, electrical current, and/or local specific heat exceeds some critical value and may lead to the flux jumps. Thus, we deduce that the only mechanism that could give rise to the flux jumps are the increase in temperature due to the Joule heat $(dQ/dt=EJ)$ induced by the temporal variation in the external magnetic field, where the vortices move as a dendritic flux (in cascade) by the interdiffusion of the local microscopic regions. This process increases the local temperature even close to Tc in some region of the sample. For instance, when the sample is irradiated by electrons or gammas, where the crystalline disorder is deeper, the flux jumps are recorded up to 23.5 K. The high critical current density, low heat capacity at low temperature, and the increases in crystalline defects in many cases like this, promote the flux jumps in the magnetization hysteresis loop and likely as avalanches growing inside the MgB_2 bulk sample. Thus, enhance either the current density, Jc or superior critical fields, H_{c2} or of both at the same time using irradiated high dense samples, external additives as nanodoping particles,[3,21] as well as epitaxial thin films in many cases produces flux jumps instabilities in MgB_2, which should be taken into consideration as disadvantages for large-scale applications and future electronic devices.

IV. CONCLUDING REMARKS

The introduction of pinning center by means of low dosages of irradiation by γ-rays, proton, and electron irradiation was investigated in order to increase the critical current density. The irradiation dosages have no effects with the critical temperature. All samples studied showed magnetothermal instabilities as flux jumps in the hysteresis loop curves. The flux jumps depend on the kind of irradiation ions and temperature. We believe that the close connection between the heat dissipation, flux lines motions, and the high density of defects trigger off magnetothermal instabilities. Nevertheless, it is observed that the irradiation process improves the field dependence of critical current density through modest levels of atomic disorder and likely redistribution of the local and superficial defects acting as effective pinning center.

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