Title

$\Delta H = \Delta B$ section in volume defect dominating superconductor.

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Abstract

According to G-L theory, it has been generally accepted that the diamagnetic property decreases after $H_{c1}$. However, we found that (Fe, Ti) particle doped MgB$_2$ specimens reveal the $\Delta H = \Delta B$ section in the $M$-$H$ curves, which are not following the G-L theory. We present whether this phenomenon appears to be only confined to (Fe, Ti) particle doped MgB$_2$ superconductor, whether there is a theoretical basis and why it does not appear in other superconductors. We have understood that the cause of $\Delta H = \Delta B$ section is the pinning phenomenon of defects in the superconductor and it only occurs in volume defect dominating superconductors. The width of $\Delta H = \Delta B$ section along the number of defects and $H_{c2}$ was estimated assuming that defects are in the ideal state, and compared with experimental results. We hypothesized that pinned fluxes have to be picked out from the defect and move into an inside of a superconductor regardless $\Delta G_{\text{defect}}$ if the distance between fluxes pinned at the defect is equal to the one of $H_{c2}$. It is considered that the reason that this phenomenon has not been reported yet is the flux jump of the volume defect dominating superconductor. The section means that the fluxes that have penetrated into a inside of a superconductor in which volume defects exist are preferentially pinned on them over the entire specimen before G-L behavior. If the size of volume defects is uniform in some extent, the influence of the planar and line defects is small and the flux jump does not occur, we believe that the section must be observed in any superconductor. It is because this is one of the basic natures of pinning phenomenon in the volume defect dominating superconductor.

MAIN TEXT

Introduction

Although it is clear that superconductors have a flux pinning effect, the exact mechanism is still disputing [1-4]. All of superconductors have flux pinning effects since they have defects even if defects are few. Most of superconductors show that the diamagnetic property of the superconductor gradually decreases after $H_{c1}$ (not $H_{c1}'$) according to Ginzburg-Landau (G-L) theory. However, we found a phenomenon in (Fe, Ti) particle doped MgB$_2$ specimens which is not following the G-L theory after $H_{c1}'$, that is the existence of the $\Delta H = \Delta B$ section in the magnetization-applied field ($M$-$H$) curves. This is an unusual phenomenon that did not reported in other superconductors.

The flux pinning phenomenon is mainly caused by defects in the superconductor. Generally, defects in the superconductor contain volume ones (such as general volume defects, precipitators, inclusions and columnar defects etc.), planar ones (such as grain boundaries, twin boundaries and stacking faults etc.) and line ones (like dislocations). Even though they all belong to a family of
defect, a role difference for the flux pinning is considerable. In the case of volume defects, pinned fluxes are hard to escape from defects except a force balance \(F_{\text{pinning}} = F_{\text{pickout}}\) being broken, hence they are called strong pinning sites.

However, weak pinning sites such as planar defects and line defects are entirely different from the strong pinning sites in flux pinning mechanism. Looking for the grain boundary (GB) which have relatively lower pinning energy caused by planar characteristic, they are connected each other in the whole specimen as a planar defect. Thus fluxes pinned on the GB move easily along GBs. In addition, since total area as a defect is large, they have a significant importance in the overall flux pinning effects. A superconductor dominated by planar defect in flux pinning effect can be called a planar defect dominating superconductor. High \(T_c\) superconductor (HTSC) bulks are associated in this category [5-8]. Since dislocations as a line defect are also interconnected throughout a specimen, a penetration of fluxes is easy through them like planar defect dominating superconductor. Worked NbTi superconducting wires are associated in this category [10, 11]. Therefore, planar and line defect dominating superconductors superficially behave like following the G-L theory.

\(\text{MgB}_2\) which was made by synthetic method in the high temperature also have grainboundaries, but most of them are low angle ones due to their fabricating characteristic [12-16]. Thus it has significantly fewer weak links than HTSC bulks produced by the solid state reaction method. Therefore, it can be called a volume defect dominating superconductor. \(\text{MgB}_2\) has been known as a superconductor which field dependence is weak, but a definite effect can be obtained by adding artificial defects because it is a volume defect dominating superconductor [13, 17].

A pure \(\text{MgB}_2\) and (Fe, Ti) particle doped \(\text{MgB}_2\) specimens for the study were synthesized using the nonspecial atmosphere synthesis (NAS) method [18]. All of the specimens which had been synthesized at 920°C for 1 hour were cooled in air, but 5 wt.% (Fe, Ti) particle doped \(\text{MgB}_2\) specimen which had shown prominent results underwent two different cooling processes. The one was cooled in air and the other was quenched in water. Figure 1 (a) is the draft of the non-special atmosphere synthesis (NAS) method for \(\text{MgB}_2\) and Fig. 1 (b) is a photograph of the method. Figure 1 (c) is a photograph of (Fe, Ti) particles, which are a little far from sphere and Fig. 1 (d) shows (Fe, Ti) particles that is present in \(\text{MgB}_2\). The radius of particles is rather irregular, and the average radius of them is 163 nm. The distribution of (Fe, Ti) particles in \(\text{MgB}_2\) are not that of the ideal one.

**Results**

A diamagnetic property increase and a confirmation of the \(\Delta H = \Delta B\) section in experiments

Fluxes would penetrate into the superconductor in a flux quantum form over \(H_{c1}\) [20]. According to the G-L theory, the diamagnetic property of the superconductor gradually decreases if fluxes have penetrated into it, and this phenomenon continues to \(H_{c2}\). This is true if there are no defects which are pinning sites in the superconductor. But real superconductors which have defects behave differently. Fluxes having penetrated into the superconductor are pinned at defects near the surface and the diamagnetic property increases rather than the one of \(H_{c1}\). We call it \(H_{c1}'\), which represents the field of the maximum diamagnetic property in the real superconductor.

In planar defect dominating superconductors and line defect dominating
superconductors, the diamagnetic property of $H_{c1}'$ did not make a great difference from the one of $H_{c1}$. It is caused by the fact that a small volume of a individual defect induces a weak pinning force. Hence the increase of the diamagnetic property at $H_{c1}'$ is small. Especially, they are interconnected, thus they allow well for a flux penetration. Therefore, it seems superficially to follow the G-L theory and there is no the $\Delta H = \Delta B$ section in $M-H$ curves.

However, volume defect dominating superconductors show a distinctly different behavior. The pinning effect is strong owing to their relatively larger volume, and the most important thing is that they are not interconnected each other. Therefore, they continue to pin fluxes until they exceed their pinning limits. They would act as another barrier to prevent fluxes from penetrating into the superconductor over $H_{c1}$. Thus, the diamagnetic property of the volume defect dominating superconductors certainly increases. As shown in all $M-H$ curves except pure MgB$_2$ in Fig. 2, a linear region ends about 600 Oe, which means a perfect diamagnetism. And after that, they show a slight decrease in slope. This means that fluxes penetrated into the superconductor are pinned at defects near the surface and cannot easily move into the specimen. Therefore, the diamagnetic property of the specimen continues to increase even though $H_{c1}$ has passed.

Figure 2 (a) are $M-H$ curves of pure MgB$_2$ and 5 wt.% (Fe, Ti) particles doped MgB$_2$ that were air-cooled and measured at 5 K. The $M-H$ curve of pure MgB$_2$ used as reference. It is clear that the $\Delta H = \Delta B$ section can be seen after $H_{c1}'$ in 5 wt.% (Fe, Ti) particles doped MgB$_2$. The width of the section can be disputed, but it is definite that the $M-H$ curve of the specimen forms the section from the $H_{c1}'$ to 8 kOe. After the flux jump, it continues to show the $\Delta H = \Delta B$ section up to 15 kOe. And it shows gradual decrease of diamagnetic properties which are the $\Delta H > \Delta B$ section over 15 kOe according to G-L theory.

And Figure 2 (b) shows the $M-H$ curve of 5 wt.% (Fe, Ti) particles doped MgB$_2$ that was water-quenched and also measured at 5 K. Generally, water-quenching method is the one to refine the grain by impeding the growth rate of grains or to induce rapid phase transformation (e.g.: martensite transformation). In this experiment, it was used for increasing the angle between grains of MgB$_2$ and refining grains due to rapid cooling rate. This treatment has the purpose of providing further opportunities for the fluxes that have been pinned at volume defects to leak out through the grain boundary. Thus this procedure can reduce the stress of fluxes concentration on the volume defects. Therefore, the $\Delta H = \Delta B$ section in the figure is formed up to 20 kOe in wide view even though there was a small flux jump. As we have seen in these two figures, it is reasonable that the $\Delta H = \Delta B$ section of 5 wt.% (Fe, Ti) particles doped MgB$_2$ specimen is from $H_{c1}'$ to a point between 15 kOe and 20 kOe. Figure 2 (c) is $M-H$ curve measured at 10 K on the air-cooled 5 wt.% (Fe, Ti) particles doped MgB$_2$ specimen and (d), (e), (f) in Fig. 2 are ones measured at 5 K with different doping concentrations. It is clear that there is the $\Delta H = \Delta B$ section in the specimens except 1 wt.% doped one.

**Pinned fluxes movement and a basis of the $\Delta H = \Delta B$ section**

It has been experimentally represented a long time ago that flux quanta pinned at the
defect move with bundle and hop from one pinning site to another pinning site [21, 22]. If the distance between the volume defects is wide enough, fluxes that are pinned at the defect do move when the force balance is broken ($F_{\text{pinning}} < F_{\text{pickout}}$), which is based on the $\Delta G_{\text{defect}}$ and a repulsive force between flux quanta. However, when the distance between the volume defects is short, we found that fluxes pinned at the defect would move into the inside of the superconductor by different mechanism. It is that they have to be picked out from the defect and move into the inside of the superconductor regardless of $\Delta G_{\text{defect}}$ when the pinning sites (defects) of the superconductor reach the limit value of pinned fluxes.

If a volume defect existing near a surface of the superconductor pins fluxes which have penetrated into the superconductor over $H_{c1}$ and pinned fluxes are blocked from moving into the inside of the superconductor until defect's pinning limit, the free energy density of a spherical defect in the superconductor is

$$\Delta G_{\text{super}} - \Delta G_{\text{nor}} = \frac{H^2}{8\pi}, \quad \Delta G_{\text{defect}} = -\frac{H^2}{8\pi} \times \frac{\pi r^2}{3},$$

(1)

Where $H$ is the applied field, $r$ is radius of defect. According to the equation, $\Delta G_{\text{defect}}$ is entirely dependent on the external field $H$ when $r$ is constant. Flux quanta pinned at the defect can move into a inside of the superconductor when they are beyond pinning limit of the defect, and they will be pinned again at another defect in front of them. It is necessary to raise $H$ in order for fluxes to be picked out from the defect and move into a inside of the superconductor. If $H$ is raised, $\Delta G_{\text{defect}}$ in the superconductor becomes larger. Thus, stronger $H$ will be needed for penetrating fluxes into a inside of the superconductor. Then $\Delta G_{\text{defect}}$ in the superconductor will continue to become larger if there are many pinning sites (volume defects).

However, although a diamagnetic property of the superconductor increases due to the pinning phenomenon, it does not increase continuously. There must be the limit value of the pinned fluxes that brakes this premise as shown in the experiment. We considered the basis of this limit to be the minimum distance between pinned fluxes at the defect. This is because the neighborhood around the defect which have pinned fluxes is no longer a superconducting state when the minimum distance between fluxes pinned at the defect is less than the one of $H_{c2}$, thus there is no pinning effect anymore. We estimated that this limit is 10.17 nm in MgB$_2$, which is the distance between fluxes at $H_{c2}$ that is 20 Tesla (T) [23]. This means that fluxes pinned at the defect have to move regardless of $\Delta G_{\text{defect}}$ when the minimum distance between fluxes pinned at the defect is reduced to 10.17 nm.

The reason of creating the $\Delta H = \Delta B$ section in the M-H curve is originated from the flux pinning limit value of a volume defect in the superconductor. It is clear that the $\Delta H = \Delta B$ section is formed in the M-H curve because defects are filled with flux quanta step by step from the surface to the center of the superconductor when defects have pinning limit of flux quanta according to its radius. Figure 3 (a) and (b) show the flux pinning limit of defects for having the same and a different radius, respectively. Both are superconductors with four defects along the y-axis and nine defects in the x-axis. The difference between the two is the particle size of defects, which is uniform in (a) and the average particle size in (b) is the same as the one of (a). Assuming that the flux quantum lies in the y-axis and
moves in the x-direction, it is natural that the $\Delta H = \Delta B$ section is formed in (a) because fluxes coming from the outside of the defect are pinned at the defect and move if a defect exceeds its flux pinning limit.

However, (b) behaves a little differently. Fluxes pinned at a small pinning site move first because the flux pinning limit is low, and fluxes pinned at larger defect move later. Since many fluxes that have been pinned at larger defect move together in this case when they pick out from the defect, there is a high possibility of flux jump. 5 wt.% (Fe, Ti) particles doped MgB$_2$ is an example of defect's unevenness which is shown in Fig. 2 (a). It has approximately $8000^3$ defects where radius is 163 nm on average in 1 cm$^3$ of MgB$_2$. In this situation, a single quantum flux in a superconductor would be pinned at 8000 defects on average. Thus there are some fluxes pinned at a defect which move first, and some fluxes at another defect which move later partially, but the ability to pin fluxes on average is similar to the counterpart when it is seen as a whole specimen. Therefore, there is no problem in forming the $\Delta H = \Delta B$ section because the number of fluxes that can be pinned on defects is predefined as a whole and they are pinned on the defects which are located from the surface to the center of the superconductor.

**Calculations for a flux pinning limit of a defect and the width of $\Delta H = \Delta B$ section**

If it is assumed that volume defects are spherical, its size is constant, and it is regularly arranged in a superconductor, a superconductor of 1 cm$^3$ has $m^3$ volume defects. The maximum number of flux quanta that can be accommodated at a spherical defect of radius $r$ in a static state is

$$n^2 = \frac{\pi r^2}{\pi(\xi/2)^2} \times P = \left(\frac{2r}{\xi}\right)^2 \times P$$

(2)

where $r$, $\xi$ and $P$ is radius of defect, coherence length and filling rate which is 0.78, respectively (see Fig. 3 (c) and (d)). If the radius of a defect is 163 nm, the maximum number of quantum fluxes that can be pinned by the defect is about $29^2$ ones at 0 K in a static state when the coherence length is 10.17 nm. We thought that quantum fluxes had a square structure rather than a triangle one when they were pinned at the defect [24].

Therefore, the magnetic induction $B$ is

$$B = n^2 m_{cps} m \Phi_0$$

(3)

where $n^2$, $m_{cps}$, $m$ and $\Phi_0$ are the number of quantum fluxes pinned at a defect, the number of defect which is vertically closed packed state, the number of defects with pinned fluxes from the surface to the center of the superconductor and flux quantum, respectively. The $m_{cps}$ is explained in Fig. 3 (e) and (f). The $m_{cps}$ is the minimum number of defects if fluxes penetrated into the superconductor are completely pinned. This conversion is introduced for calculating the number of flux quanta which are pinned on defects of a plane since fluxes between the defects can penetrate into the superconductor without pinning if defects are perfectly arranged like a lattice. In reality, defects are randomly arranged in the real superconductor. If $8000^3$ defects are in 1 cm$^3$ superconductor as described in the
experiment, there are approximately $8000^2$ defects in a plane. Thus, there are almost no penetrating fluxes without pinning. Thus, the total numbers of flux quanta pinned on the defects of a plane perpendicular to the flux moving direction are $n^2m_{\text{cps}}$.

Therefore, the magnetization $M$ is

$$ B = H + 4\pi M, \quad M = \frac{n^2m_{\text{cps}}}{4\pi} \left( \Phi_0 - \frac{H}{4\pi} \right) $$  \hspace{1cm} (4)$$

If a radius of the defects is fixed, $n^2$ and $m_{\text{cps}}$ is also fixed. Thus the width of the $\Delta H = \Delta B$ section is only dependent on the $m$. A calculated width of the $\Delta H = \Delta B$ section along a number of defect is shown in Fig. 4 (a) assuming that the radius of a defect is 165 nm and $H_{c2}$ is 20 T. As shown in the figure, the more pinning sites are, the wider the $\Delta H = \Delta B$ section is except over-doping.

One of important factors calculating the width of $\Delta H = \Delta B$ section is what is $H_{c2}$ of MgB$_2$. $H_{c2}$ is the fundamental property according to a material of superconductor, but it is inferred from an indirect method at a low temperature since it has difficulty to be directly measured. Thus, $H_{c2}$ of MgB$_2$ also varies from 64 T of theories to about 20 T of experiments according to workers [22, 25-27]. A calculated width of the $\Delta H = \Delta B$ section along $H_{c2}$ variation is shown in Fig. 4 (b) assuming that the radius of a defect is 165 nm and there are $8000^3$ defects in 1 cm$^3$ superconductor which is equivalent to 5 wt.% (Fe, Ti) particle doped MgB$_2$. When the width of $\Delta H = \Delta B$ section is calculated with $H_{c2} = 20$ T, it closely matches the experimental results as shown in the Fig. 4 (a) and (b), which were converted from 5 K to 0 K (the width of $\Delta H = \Delta B$ section was determined to be 1.7 T at 5 K). The difference between the calculated value and the experimental value in the figure is considered to be the result of the fact that our experiments were not carried out in ideal conditions. Figure 4 (c) shows the flux penetration method based on the G-L theory and (d), (e) and (f) show the flux penetration method based on the existence of the $\Delta H = \Delta B$ section. They indicate that the fluxes penetrated into the superconductor are pinned preferentially on the volume defects over the entire specimen before G-L behavior. The width of $\Delta H = \Delta B$ section increases as the number of volume defects increase, and the width of the section is narrow if the number of volume defects a few and too many.

**Discussion**

MgB$_2$ has weak magnetic field dependence. In order to solve this problem, many researchers have doped a variety of materials and achieved considerable results [28-30]. However, despite improved field dependence in high field, there were still a lot of flux jumps in MgB$_2$. We have thought that it is caused by the fact that MgB$_2$ is a volume defect dominating superconductor. Thus it is considered that MgB$_2$ researchers overlooked the existence of $\Delta H = \Delta B$ section in their specimens. The essence of this paper is that fluxes penetrated into the superconductor does not follow the G-L theory and are preferentially pinned on defects over the entire specimen if it is volume defect dominating superconductor. This is because fluxes pinned on the defect are bent like a bow, thus unpinned ones are hard to exist without pinning due to the repulsive force between fluxes and irregular distribution of defects. Since the external field has already exceeded $H_{c1}$,
internal fluxes (B) increase as much as the external field increases. Therefore, when the superconductor is volume defect dominating one, $\Delta H = \Delta B$ section is first formed after $H_{c1}$ and then $\Delta H > \Delta B$ section is formed later in the M-H curve.

On the other hand, many superconducting researchers are hard to accept that flux pinning on defects cause a larger diamagnetic property than the one of $H_{c1}$. But this is a common phenomenon because there is no material having no defects. It is rather natural to explain that planar and line defect dominating superconductors follow the G-L theory is due to the interconnectivity of the defects. A typical example of increasing a diamagnetic property by flux pinning is the fishtail effect. The fishtail effect is often seen in superconducting single crystal (SC), especially in HTSC SCs. There are many opinions about the cause of the fishtail effect, but there is a consensus that it is due to the pinning phenomenon [31, 32].

One of important features of volume defect dominating superconductor (VDS) is the flux jump. If pinned fluxes on volume defects do not leak out through planar defects, they will pin fluxes to their pinning limit. And they move together when they are picked out from the defect, thus flux jump may occur if they are many. Moreover, since diamagnetic property of the VDS is always higher than that of the pure state of superconductor and the superconductor with volume defects are pinned from the surface, fluxes pinned on defects are always under the pressure that they may penetrate into the inside of the superconductor. This is the reason that the flux jump occurs well in MgB$_2$ which are synthesized by Mg and B. The basic reason that a $\Delta H = \Delta B$ section has not been reported so far is considered to be a lack of a proper VDS like MgB$_2$. And although VDSs were made as single crystals and unworked NbTi etc., it could not be observed due to the flux jump.

Other examples of VDS are single crystals (SC) of superconductor and melt-texture growth (MTG) specimens of HTSC. In both cases, the superconductor is formed in the melted state, so the influence of planar defect and line defect are weak [33, 34]. One may say what volume defects are in a single crystal, but there are many volume defects in it. Assuming that base materials of 99.9999% purity are used to make a single crystal, one of a million of atoms is a impurity in the SC. Calculated roughly, if there are $1 \times 10^{23}$ atoms in $1 \text{ cm}^3$, the number of impurities are $1 \times (10^9)^3$ ones. This means that about $1 \times 10^6$ defects exist along one axis. Although they exist in the form of inclusion that impurity atoms exist in a dense form of about 1000 atoms, there are $1 \times 10^3$ volume defects along one axis. Especially, even if only one impurity atom enters a unit lattice in case of HTSC SCs, the unit lattice becomes a volume defect. Thus though effects of volume defects in SC is small compared with polycrystals, they must not be ignored. In addition, SC superconductors often contain inevitable planar defects due to its material characteristics of primitive lattice. Two cases are representative, which are stacking faults due to the interstitial nature of boron in MgB$_2$ and twin boundaries that were caused by phase transformation during cooling from tetragonal into rhombic structure in HTSCs, which have a direction in contrast with GBs [35-37].

The fewer planar defects are, the higher a dominance of volume defects is. However, since the concentration of impurity is low in SC superconductors, the number of volume defects is a few. Thus the width of $\Delta H = \Delta B$ section will be short as shown in Fig. 4 (a).
HTSC SCs generally show the flux jump instead of showing the section in the M-H curve when twin boundaries are few [38]. However, if there are twin boundary plane which is perpendicular to the moving direction of fluxes, (if the longitudinal direction of the fluxes and the twin boundary plane are parallel), strong pinning effects occurs. And if there are many twin boundaries in SC which is not perpendicular to the moving direction of fluxes, fluxes that have been pinned to volume defects leak out along planar defects and the M-H curve behaves just like G-L theory [39, 40].

On the other hand, the stacking fault planes formed on the MgB2 SC are not directional. Thus M-H curves of it are similar to the one of an ideal state that a pinning effect does not occur at all. It is considered that this behavior is because the fluxes move easily along the stacking fault planes. Y. Machida et al. reported that Tc was 38 K which is rather lower than that of polycrystalline specimen (39 K) [35]. And they suspected the contamination of specimen but had no results. On the other hand, M. Xu et al. also reported M-H curve of MgB2 SC which is similar with the one of Y. Machida et al. and the composition of MgB2 SC was determined to be MgB1.9 [36]. When above results are considered with the other fact that there is a large stress between Mg and B, which is caused by the crystal structure that B is interstitial element in MgB2, it is speculated that the stress between Mg and B released by stacking faults in MgB2 SC. If the ratio of Mg and B is 1 : 1.9, a layer of 30 ones in the SC is statistically a stacking fault plane. Therefore, fluxes in the MgB2 single crystal move easily along the stacking fault plane, thus the MgB2 SC shows very low hysteresis in M-H curve.

MTG was introduced to eliminate the HTSC's weak links. One of the distinct features of MTG is that flux jump occurs frequently like MgB2. The fact that the flux jump, which was not observed in HTSC specimen prepared by the solid-phase reaction method, frequently occurs in HTSC specimen prepared by the MTG method means that the dominating flux pinning mechanism has changed from planar defects to volume defects. Since MTG has a high concentration of impurities compared with SC, we searched hard for various papers in the hope that there would be the section in M-H curves of MTG and found two papers [41, 42]. We can see that a considerable width of \( \Delta H = \Delta B \) section has been formed after \( H_{c1} \) in these papers. Therefore, we have clear confirmation for that the phenomenon which \( \Delta H = \Delta B \) section appears is not to be confined to MgB2, especially this experiment but to be common in volume defect dominating superconductors.

We proved the \( \Delta H = \Delta B \) section by the experiments of (Fe, Ti) particles doped MgB2 specimens that are a volume defect dominating superconductor. And we postulated a new concept that this behavior of the superconductor is based on the flux pinning limit of defects regardless of the \( \Delta G_{\text{defect}} \). And it was also emphasized that superconductors should be classified as not materials but defects in order to understand the flux pinning phenomena properly. There is no defect-free material, and it is proper to interpret the phenomenon of superconductivity based on this point. We think that the \( \Delta H = \Delta B \) section is the basis of the flux pinning phenomenon which is shown ahead of G-L behavior in volume defect dominating superconductor. It has been buried in the flux jump.

Materials and Methods
The starting materials were Mg (99.9% powder) and B (96.6% amorphous powder) and (Fe, Ti) particles. Mixed Mg and B stoichiometry, and (Fe, Ti) particles were added by weight. They were finely ground and pressed into 10 mm diameter pellets. (Fe, Ti) particles were ball-milled for several days, and average radius of (Fe, Ti) particles was about 163 nm. On the other hand, an 8 m-long stainless-steel (304) tube was cut into 10 cm pieces. One side of the 10 cm-long tube was forged and welded. The pellets and excess Mg were placed in the stainless-steel tube. The pellets were annealed at 300°C for 1 hour to make them hard before inserting them into the stainless-steel tube. The other side of the stainless-steel tube was also forged. High-purity Ar gas was put into the stainless-steel tube, and which was then welded. All of the specimens had been synthesized at 920°C for 1 hour. The field and temperature dependence of magnetization were measured using a MPMS-7 which was produced by Quantum Design. During the measurement, sweeping rates of all specimens were made equal for the same flux-penetrating condition.

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Figures and Tables

Fig. 1. The non-special atmosphere synthesis (NAS) method for MgB₂ and (Fe, Ti) particles for the experiment. (a): The draft of the non-special atmosphere synthesis (NAS) method. (b): A photograph of the specimen for the NAS method. (c): A photograph of (Fe, Ti) particles before doped in MgB₂, which were ball-milled for several days. (d): A photograph of 25 wt.% (Fe, Ti) particles doped MgB₂, which was taken by the field emission scanning electron microscope (FE-SEM). White bright ones in MgB₂ base are doped (Fe, Ti) particles.
Fig. 2. Field dependences of magnetization for pure MgB$_2$ and (Fe, Ti) particles doped MgB$_2$ (M-H curves). (a): A field dependence of magnetization for pure MgB$_2$ and 5 wt.% (Fe, Ti) particles doped MgB$_2$. Specimens were air-cooled and measured at 5 K. (b): A field dependence of magnetization for 5 wt.% (Fe, Ti) particles doped MgB$_2$, which was water-quenched and also measured at 5 K. (c): A field dependence of magnetization for 5 wt.% (Fe, Ti) particles doped MgB$_2$, which was air-cooled but measured at 10 K. (d): A field dependence of magnetization for 1 wt.% (Fe, Ti) particles doped MgB$_2$, which was air-cooled and measured at 5 K. (e): A field dependence of magnetization for 10 wt.% (Fe, Ti) particles doped MgB$_2$, which was air-cooled and measured at 5 K. (f): A field dependence of magnetization for 25 wt.% (Fe, Ti) particles doped MgB$_2$, which was air-cooled and measured at 5 K.
Fig. 3. The flux pinning limit of defects, filling rate calculation and the definition of $m_{\text{csp}}$. (a): The flux pinning limit of defects when they have same radius and regular arrangement. (b): The flux pinning limit of defects when they have different radius and regular arrangement. (c): The shape of the maximum fluxes pinned at the defect, which is cut off the center of the defect and assumed that it is spherical. (d): Assuming that the coherence length ($\xi$) is 10.17 nm, the maximum number of fluxes pinned at the defect of the radius $r$ can be obtained. (e): The ideal arrangement of defects. There is a possibility that fluxes are not pinned at defects if fluxes lie on the y axis and move along the x-axis. (f): The $m_{\text{csp}}$ state. The $m_{\text{csp}}$ is the number of defects which are vertically closed packed state of defects. The defect arrangement in (e) has to change to ones in the (f) in calculating B in the superconductor for not having any flux quantum penetrating into the superconductor without pinning.
Fig. 4. A calculated width of the $\Delta H = \Delta B$ section and flux penetration method compared to the G-L theory. (a): A calculated width of the $\Delta H = \Delta B$ section along the number of defects in MgB$_2$. (b): A calculated width of the $\Delta H = \Delta B$ section along a variation of upper critical field ($H_{c2}$) of MgB$_2$. (c): The flux penetration method which are based on the G-L theory [24]. (d): The flux penetration method when superconductor has a good pinning condition in volume defect dominating superconductor. (e): The flux penetration method when superconductor has a proper pinning condition in volume defect dominating superconductor. (f): The flux penetration method when superconductor has a poor pinning condition in volume defect dominating superconductor.
Supplementary Materials

Text S1. Elements that affect the width of $\Delta H = \Delta B$ section

Elements that affect the width of $\Delta H = \Delta B$ section can be expressed as follows

$$L_{\Delta H=\Delta B} = L_{\text{ideal}} - L_{\text{reduction}}$$

(S1)

where $L_{\text{reduction}} = L_{\text{unisize}} + L_{\text{distri}} + L_{\text{temp}} + L_{\text{dyn}} + L_{\text{shape}}$.

$L_{\text{unisize}}$ is the element concerning to the unevenness of the defect size. If sizes of defects is not the same, a defect of a large size will relatively pin a lot of fluxes, and a defect of a small size will pin a few fluxes. Let's looking on the effect of the $\Delta H = \Delta B$ section when the defect of radius $r$ is split into the one of $r/2$ assuming that there is $n^3$ volume defects of radius $r$ in the superconductor. The number of defects will be $2^3 n^3$ if a radius of defects becomes $r/2$. This means that two defects increase with each axis. Amount of fluxes pinned on a defect reduce to $(k/2)^2$ when its radius becomes $r/2$ if maximum fluxes that a defect of radius $r$ can pin is $k^2$. Therefore, when the radius of a defect is reduced to $r/2$, the number of fluxes pinned on defects is reduced to 1/2 along a axis and the width of the section is also reduced to 1/2 because the width of the section is the total amount of fluxes pinned on defects to an axis. Since the average radius of defects used in the experiment is 163 nm, defects which are smaller than average one would pin fluxes fewer and defects which are larger than average one would pin fluxes more, respectively. The total amount of fluxes that can be pinned on defects of the same radius is similar to the total amount of fluxes that can be pinned on defects of the same average radius when the number of defects on an axis is same. Thus, it is considered that although the difference in size of the defects will affect the $\Delta H = \Delta B$ section, it does not have a significant effect.

$L_{\text{distri}}$ is the element concerning to non-uniform distribution of defects. This refers to the effect of the differences between the ideal spacing and the non-ideal spacing of defects. Since fluxes move forward when a defect reaches its pinning limit, there is no significant effect on flux pinning limit of defect if the distance between two defects is different to some extent (if the distance between two defects is significantly different from the average one, the repulsive forces of the flux quantum reduces flux pinning limit of the defect). Furthermore, since one flux quantum is pinned at 8000 defects on average at the same time (if defects are (Fe, Ti) particles with a radius of 163 nm and 5 wt.% is contained in MgB$_2$), the effect on the width of the section by non-uniform distribution of defects is minimal.

$L_{\text{temp}}$ is the element of temperature. As a temperature rises, the fluctuation of the flux quanta become larger, and the pinning limit of a defect reduces. It is known that the coherence length ($\xi$) follows the equation
where \( t = T/T_c \) [1]. Since the coherence length is about 10.17 nm when \( H_{c2} \) is 20 T at 0 K, it becomes about 10.72 nm at 5 K. Thus, the maximum number of fluxes that can be pinned in a defect which radius is 163 nm is about \( 27^2 \) at 5 K and the width of the section is about 2 T, whereas it is about \( 29^2 \) at 0 K and the width of the section is about 2.25 T. Therefore, if the width of the section at 0 K is compared with the one at 5 K, the difference is more than 10 %.

\[
\xi(T)^2 = \frac{\hbar^2}{2m^*\alpha(T)} \propto \frac{1}{1 - t}
\]

\text{(S2)}

\( L_{\text{dyn}} \) is the element of flux pinning state. \( L_{\Delta H} = \Delta B \) in ideal condition was calculated under a static state of magnetic field. When flux quanta come from an outside of the defect, they are moving and not in the stable position. It is certain that fewer fluxes will be pinned compared with ones under the static state if they are under a dynamic state. However, it is considered that there are some kinds of fluxes which are not pinned on defects during movements of fluxes. When they reach a static state, they are difficult to exist between defects due to repulsive forces with the pinned fluxes on defects and bending shape of them. They will be pinned on the defects which can afford to pin fluxes more under a static state, and the remaining flux quanta which are unpinned will be pushed forward. For this reason, it is expected that the number of flux quanta in the superconductor under dynamic and static states will not show much difference. \( L_{\text{shape}} \) is the element of defect’s shape. According to the shape of the defect, the pinning limit may be changed. A spherical shape of defect is the safest one, thus they can pin the greatest number of quantum fluxes. The larger the degree of deviation from spherical shape is, the less the amount of fluxes can be pinned. (Fe, Ti) particles which were used for artificial pinning sites (APT) is not a perfect spherical, but it does not seem to be too far away from a sphere. Though the effect of defect’s shape is, it is not prominent.

As a result of examining various elements influencing a width of the section in experiments, the influence of temperature is the most prominent, and rests are considered not to be much larger than the influence of temperature. Calculating results were compared with the experimental data, the two matched well with a relatively small difference. Experimental data of Fig. 4 (a) and (b) were converted from 5 K to 0 K. The difference between the calculated value and the experimental one in figures seems to be the remaining value of \( L_{\text{reduction}} \) except \( L_{\text{temp}} \).

**Text S2. Another evidence that the water-quenched specimen will leak out more easily the pinned fluxes on volume defects**

Figure S1 shows the M-H curve by reducing the sweep rate of 5 wt.% (Fe, Ti) doped MgB\(_2\) which was air-cooled. And the inset shows the M-H curve of water-quenched 5 wt.% (Fe, Ti) doped MgB\(_2\). The sweep rate of the air-cooled specimen was reduced to 1/3 compared to Fig. 2 (a). Two figures are rather similar each other except high field region around 5 T (especially, the fact that the flux jump is reduced and the angle between the line of \( \Delta H = \Delta B \) section and the line of horizon). To
reduce the sweep rate to 1/3 tells us that fluxes pinned at volume defects have three times longer than the counterpart to leak out through the grain boundary. Thus the fact that the M-H curve of reducing the sweep rate to 1/3 is the same as one of a water-quenched specimen means that water-quenched specimen leaked out fluxes pinned at the defect three times faster than air-cooled specimen did.

**Fig. S1.**: Field dependence of magnetization along the magnetic field of 5 wt.% (Fe, Ti) doped MgB$_2$. The sweeping rate was reduced to 1/3 compared to Fig. 2 (a). The inset shows the M-H curve of water-quenched 5 wt.% (Fe, Ti) doped MgB$_2$.

[1] Michael Tinkham, *Introduction to superconductivity* (Dover Publication, New York, 2004). [second edition], p. 118.