Flux-pinning mechanism in silicone-oil doped MgB$_2$: Evidence for charge-carrier mean free path fluctuation pinning

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Abstract
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Keywords
Flux, pinning, mechanism, silicone, oil, doped, MgB2, Evidence, for, charge, carrier, mean, free, path, fluctuation, pinning

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Flux-pinning mechanism in silicone-oil-doped MgB$_2$: Evidence for charge-carrier mean free path fluctuation pinning

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Flux-pinning mechanism of MgB$_2$ doped with 10 wt % silicone-oil sintered at low and high temperatures has been investigated by magnetic measurements. The field dependence of the critical current density, $j_c(B)$, was analyzed within the collective pinning model. A crossover field, $B_{c0}$, from the single vortex to the small vortex bundle-pinning regime was observed. For both types of sintered samples, the temperature dependence of $B_{c0}(T)$ at low temperature is in good agreement with the $\delta B$ pinning mechanism, i.e., pinning associated with charge-carrier mean free path fluctuation. At temperatures close to the critical temperature, however, there is evidence for $\delta T_c$ pinning, which is associated with spatial fluctuations of the transition temperature. These results provide strong evidence that the liquid precursor, silicone oil, produces very small pinning centers and enhances the $j_c(B)$.

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I. INTRODUCTION

Magnesium diboride MgB$_2$ has the highest superconducting transition temperature ($T_c \approx 39$ K) among the metallic compounds. The critical current density has been a central topic of research since discovery of superconductivity in this compound. High critical current-density values of $10^5$–$10^6$ A/cm$^2$ have been reported for this superconductor. However, the critical current density drops rapidly with increasing magnetic field due to its poor flux pinning. At the irreversibility field, $H_{irr}$, vortices start to move along the direction of the current flow, and hence the critical current vanishes. The current-density decay behavior is governed by the pinning mechanism. Numerous studies have been performed with the purpose of understanding the vortex-pinning mechanisms that are responsible for improving the critical current density, $j_c$. Intergrain boundary pinning and point defect pinning are two main important pinning mechanisms.

In type-II superconductors, the most important elementary interaction between vortices and pinning centers is the magnetic interaction and the core interaction. The core interaction arises from the coupling of the locally distorted superconducting properties with the periodic variation in the superconducting order parameter, which is usually more effective in type-II superconductors due to the high $k$ value. It has been found that in MgB$_2$ bulk and thin-film samples, with $\kappa$ larger than 20, the core pinning is related to randomly distributed spatial variations in the transition temperature ($\delta T_c$ pinning). However, in the high-temperature superconductors the core interaction is associated with charge-carrier mean free path variations ($\delta B$ pinning), mostly due to crystal lattice defects.

The critical current density of MgB$_2$ can be improved by more than 1 order of magnitude in high magnetic field by adding nano-SiC. It has also been found that maximum flux-pinning forces are increased by decreasing grain size. One way to decrease the grain size is to use a liquid precursor, silicone oil, which can produce Si and C at the atomic scale. We have found that a significant flux-pinning enhancement in MgB$_2$ can be easily achieved in this way. Our results showed that the Si and C released from the decomposition of the silicone oil formed Mg$_2$Si and substituted into B sites, respectively. The inclusion of the silicone oil leads to a reduction in the lattice parameters, as well as $T_c$ and the relative resistance ratio (RRR) $[R(300 \text{ K})/R(T_c)]$ resulting in a significant enhancement of $J_c(H)$, $H_{irr}$, and $H_{c2}$.

Due to the defects and nanoparticle inclusions related $J_c$-field enhancement in the silicone-oil-doped MgB$_2$, the pinning mechanisms are of interest in such samples from the point of view of both the fundamental physics and applications. However, it has not been experimentally determined whether the $\delta T_c$ pinning or the $\delta B$ pinning is the dominant mechanism in MgB$_2$ when it is doped on the atomic scale.

In this paper, the vortex-pinning mechanisms of silicone-oil-doped samples are discussed in the framework of the collective theory. It was found that charge-carrier mean free path fluctuation pinning ($\delta B$) is the only important pinning mechanism at low temperature, while $\delta T_c$ pinning is effective at $T$ close to $T_c$.

II. EXPERIMENTAL

Polycrystalline MgB$_2$ samples with 10 wt % silicone-oil addition were prepared by the standard solid-state powder processing technique, which has been well described elsewhere. Because the grain size of MgB$_2$ also plays an important role in governing the flux pinning, we fabricated silicone-oil-doped MgB$_2$ at both 600 and 900 °C with the aim of achieving small and large grains of MgB$_2$, respectively. Although x-ray diffraction (XRD) results revealed that all samples were crystallized in the MgB$_2$ structure as the
major phase, a few impurity lines of MgO and Mg2Si were observed. Magnetic and transport measurements were performed using a physical properties measurement system (PPMS) (Quantum Design). The magnetic hysteresis loops of the samples were measured over a temperature range of 20–34 K for both samples. The critical current density was calculated by using the Bean approximation, \( j_c = 20\Delta M/Va(1-a/3b) \), where \( a \) and \( b \) are the width and the length of the sample perpendicular to the applied field, respectively, \( V \) is the sample volume, and \( \Delta M \) is the height of the \( M-H \) hysteresis loop.

III. RESULTS AND DISCUSSIONS

The \( j_c(B,T) \) results for both samples sintered at 600 and 900 °C are shown in a double-logarithmic plot in Fig. 1. At 4 T and 20 K, the \( j_c \) values for both samples are over 1 \( \times 10^4 \) A/cm², more than 1 order of magnitude higher than that for pure MgB2. This is due to enhancement of flux pinning, which may be ascribed to the fine nanoparticles, mainly Mg2Si, C, and MgO, which occur as inclusions inside MgB2 grains. The \( j_c \) initially shows a plateau at low field and then begins to decrease quickly as the field reaches a crossover field, which is decreased with increasing temperature. Further increase in the field results in a faster drop in \( j_c \) near the irreversibility line, which is obtained by using the criterion of \( j_c = 100 \) A/cm², as shown in Fig. 2. For all fields and temperatures higher than 29.5 K, the critical current density of samples sintered at 900 °C is higher than those sintered at the 600 °C. This is in agreement with the higher \( B_{c2} \) (see Fig. 2) and the increased grain connectivity resulting from the higher-temperature reaction. At low magnetic fields and temperatures, samples sintered at 600 °C have higher \( j_c \) values than that sintered at 900 °C. This results is surprising but is consistent with the SiC-doped MgB2. Grain boundary pinning may also play a role because it is usually important at low field region as seen in undoped MgB2.

The temperature dependence of the upper critical fields, \( B_{c2} \), which is obtained from the 90% values of their corresponding resistivity transitions, are shown in Fig. 2. As typically reported, the \( B_{c2} \) curve shows a slight upward curvature close to \( T_c \) for both sintering temperatures, and it increases with increasing temperature of the reaction. The solid curves in Fig. 2 are in close agreement with the typical behavior expected in the case of giant flux creep, which is predicted from the \((1-T/T_c)^{2} \) (where \( T = T/T_c \)) behavior for \( B_{c2} \). By extrapolating the \( B_{c2} \) curve to zero temperature, we found \( B_{c2} \) (10 K) to be about 28.04 and 31.5 T for the 600 and 900 °C reaction temperatures, respectively. These results are in good agreement with \( B_{c2}(10 \) K) of 10% nano-SiC-doped bulk MgB2 samples. This fact further indicates that the silicone-oil doping is a very effective and cheap dopant which produces excellent \( j_c \) field dependence at the same level as state-of-the-art nano-SiC-doped MgB2.

The coherence length, \( \xi \), can be calculated from the \( \xi(T)=\left(\phi_0/2\pi B_{c2}(T)\right)^{1/2} \) relation at different temperatures, where \( \phi_0 \) is the magnetic-flux quantum. \( \xi(T) \) results as a function of the normalized temperature \( T/T_c \) and the reaction temperature are shown in the inset of Fig. 3. The \( \xi \) of the MgB2 sintered at 600 °C is slightly larger than that of the 900 °C MgB2. However, the decreasing trends are same for both reaction temperatures.

In order to understand the critical current-density results shown in Fig. 1, the pinning force \( F_p= F \times j_c \) was calculated. The scaling behavior for the normalized volume pinning force \( f_{p}=F_{p}/F_{p,max} \) was examined for both \( h_1=B_{irr} \) and \( h_2=B_{max}/B_{p,max} \), where \( B_{max} \) is the magnetic field at the maximum of \( F_p \). The results are shown in Fig. 3. The \( f_p(h_z) \) curve

FIG. 1. (Color online) Magnetic and temperature dependences of the critical current density \( j_c \) for 600 and 900 °C reaction temperatures. The solid curves are fits to Eq. (2).

FIG. 2. (Color online) The upper critical and irreversibility fields of the 10 wt % silicone-oil-doped MgB2 as a function of temperature for two different sintering reaction temperatures. The solid curves are fits to \( B_{irr}=B_{irr}(0)[(1-T/T_c)^{2}]^{1/2} \) with fitting parameter \( B_{irr}(0) \). Inset: The coherence length \( \xi \) of silicone-oil-doped MgB2 as function of the normalized temperature \( T/T_c \) and the reaction temperature. Open symbols: 600 °C. Solid symbols: 900 °C.
FIG. 3. (Color online) Magnetic dependence of the reduced pinning force \( f(h) \) at a temperature range of 20–34 K. (a) \( h_2 = B/B_{\text{max}} \) for the 900 °C sintered sample, (b) \( h_3 = B/B_{\text{irr}} \) for the 900 °C sintered sample, (c) \( h_2 = B/B_{\text{max}} \) for the 600 °C sintered sample, and (d) \( h_1 = B/B_{\text{th}} \) for the 600 °C sintered sample. Solid curves are fittings to two different types of point pinning centers. The dashed curves represent curves for correlated pinning and point pinning. Therefore, we cannot obtain any dominant pinning mechanism in both reaction temperatures. At normalized magnetic fields larger than \( h_{\text{max}} \), flux pinning is dominated by correlated pinning for the sample sintered at 600 °C at temperatures lower than 34 K, while at \( T \approx 34 \) K, point pinning is dominant. For the 900 °C sample at temperatures lower than 32.5 K, the experimental data are located between the theoretical curves for correlated pinning and point pinning. Therefore, we cannot obtain any dominant pinning mechanism. In another sort of scaling behavior, the \( f_p(h_1) \) at \( T < 29.5 \) K for the 600 °C sample and \( T < 34 \) K for the 900 °C sample exhibits scaling behavior.

For the scaling, a common law of \( f_p(h_1) = A h_0^p (1-h_1)^q \) is usually employed as a single pinning function, where \( p \) and \( q \) are parameters describing the pinning mechanism. In this model, \( p = 1/2 \) and \( q = 2 \) describe normal core correlated pinning, while \( p = 1 \) and \( q = 2 \) describe normal core point pinning, as was predicted by Kramer. The best fit of the curves [solid curves in Figs. 3(b) and 3(d)] are obtained with \( p = 0.9 \) and \( q = 2.9 \) for 600 and 900 °C reaction temperatures, respectively. Therefore, we cannot infer the real dominant pinning mechanism from this scaling behavior.

As mentioned before, the \( j_s \) of both samples shows at least one crossover field, such as \( B_{sb} \). In the framework of the collective theory, which was derived by Blatter et al., the critical current density is field independent when the applied magnetic field is lower than the crossover field. In the regime below \( B_{sb} \), a single vortex-pinning mechanism governs the vortex lattice,

\[
B_{sb} \propto j_s B_{c2}.
\]

where \( j_s \) is the critical current density in the single vortex-pinning regime. At higher fields, for \( B > B_{sb} \), \( j_s(B) \) decreases quickly, and it follows an exponential law,

\[
-j_s(B) = j_s(0) \exp[-(B/B_0)^{3/2}],
\]

where \( B_0 \) is a normalization parameter of the order of \( B_{sb} \). For \( B > B_{th} \), a power dependence in the form of \( j_s(B) \propto B^{-\beta} \) acts from \( B_{th} \) to another crossover field \( B_{th} \) (large bundle-pinning regime).

The experimental results shown in Fig. 1 for \( j_s(B) \) can be described in terms of Eq. (2). The solid curves in Fig. 1 illustrate the fits to the collective model according to Eq. (2) with fitting parameters for \( j_s(0) \) and \( B_{th} \). For clarification, \(-\log[j_s(B)/j_s(0)] \) as a function of \( B \) is shown in a double-logarithmic plot in Fig. 4. It is clear that Eq. (2) well describes the experimental data for intermediate fields, while deviations from the fitting curves can be observed at both low and high fields. The deviation at low fields, below \( B_{sb} \), is associated with crossover from the single vortex-pinning regime to the small bundle-pinning regime. The high-field deviation that is very close to the irreversibility line could be related to large thermal fluctuations, a view that is supported by the three-dimensional (3D) flux creep dependence observed for the variation in \( B_{th}(T) \) in Fig. 2. The field of this deviation is denoted as \( B_{sb} \). The large-bundle pinning does not fit our data. Above this field, vortex will become very soft and then \( H-T \) diagram will enter to the very narrow
The crossover field $B_{sb}$ as a function of temperature is shown in Fig. 5 for the samples sintered at 600 and 900 °C. As can be seen in Fig. 5, the crossover field $B_{sb}$ is almost the same for both reaction temperatures. Therefore, $B_{sb}$ is not sensitive to the reaction temperature. However, the $B_{sb}(T)$ is dependent on the reaction temperature and increases with increasing sintering temperature (see Fig. 7).

Griessen et al. pointed out that the $\delta T_c$ and $\delta l$ pinning mechanisms result in different temperature dependencies of the critical current density $j_c$, in the single vortex-pinning regime. They found that $j_c \propto (1-T^2)^{3/2}(1+T^2)^{5/2}$ with $t=T/T_c$, for the case of $\delta T_c$ pinning, while for $\delta l$ pinning, $j_c \propto (1-T^2)^{3/2}(1+T^2)^{-1/2}$. Inserting these two $j_c(T)$ expressions into Eq. (1), the following temperature dependence for $B_{sb}$ can be obtained:

$$B_{sb}(T) = B_{sb}(0) \left(1 - \frac{T^2}{1 + T^2}\right)^{\nu},$$

where $\nu=2/3$ and 2 for $\delta T_c$ and $\delta l$ pinning, respectively. In the inset of Fig. 5, the dashed curves indicate the $\delta T_c$ and $\delta l$ pinning mechanisms, respectively. The curve has a positive curvature in the $\delta T_c$ pinning case, while the curvature associated with the $\delta l$ pinning is negative. As is clear from Fig. 5, the $B_{sb}(T)$ behavior shows a negative curvature. There is also a good agreement between our experimental points and Eq. (3) with $\nu=2$, and that strongly suggests that the $\delta l$ pinning mechanism is dominant, especially in low temperatures.

To investigate further the real pinning mechanism of the silicone-oil-doped MgB$_2$ samples, the $B_{sb}$ data was analyzed within the following expression:

$$B_{sb} = P_1B_{sb}^{T} + P_2B_{sb}^{l},$$

where $B_{sb}^{T}$ and $B_{sb}^{l}$ are the expression for $\delta T_c$ and $\delta l$ pinning, respectively. $P_1$ and $P_2$ are fitting parameters with $P_1+P_2 = 1$. The $B_{sb}$ data obtained from $j_c(B)$ was well described by Eq. (4), as shown by the solid curves in Fig. 5. In order to compare the effects of the $\delta T_c$ and the $\delta l$ pinning mechanisms, the $P$ parameter was defined as $P=P_1B_{sb}^{T}/B_{sb}$ or $P=P_2B_{sb}^{l}/B_{sb}$, which represent $\delta T_c$ or $\delta l$ pinning effects, respectively.

$\delta T_c$ and $\delta l$ pinning contributions as functions of temperature. Open symbols: 600 °C. Solid symbols: 900 °C.
respectively. The results of both pinning effect contributions are shown in Fig. 6. For both the 600 and 900 °C reaction temperatures, the trends in both $\delta T_c$ pinning and $\delta l$ pinning are the same, while the actual values of the pinning contribution are slightly different. As can be seen in Fig. 6, $\delta l$ pinning is the dominant mechanism at low temperature but with increasing temperature, $\delta l$ pinning decreases, and $\delta T_c$ pinning increases. At $T = 31-32$, both pinning mechanisms have equal effects, and above these temperatures, $\delta T_c$ pinning is the dominant.

Using the derived crossovers $B_{sb}$ and $B_{th}$, the reconstructed $B-T$ phase diagram is shown in Fig. 7. According to the collective pinning model, the disorder-induced spatial fluctuations in the solid-vortex lattice can be clearly divided into markedly different regimes according to the strength of the applied field. Three different regimes are at least distinguishable: (1) single vortex pinning, which governs the region below $B_{sb}$; (2) small bundle pinning, which holds between $B_{sb}$ and $B_{th}$; (3) the region between $B_{th}$ and $B_{irr}$ where thermal fluctuations are important; and (4) vortex liquid region. The comparison with the phase diagram of a pure MgB$_2$ sample shows quite different trends and areas of the each vortex phases. The silicone-oil-doped MgB$_2$ samples show very wide regions of the vortex phases, which originate from the vastly enhanced vortex pinning. Especially at low temperature, each curve exhibits upward curvature which indicates even stronger pining behavior at low temperature.

In conclusion, we have found that the $\delta l$ pinning mechanism due to spatial fluctuations of the charge-carrier mean free path is strongly dominant at low temperature in MgB$_2$ that was doped with silicone oil and sintered at both 600 and 900 °C, while at temperatures close to the critical temperature, $\delta T_c$ pinning is effective. The field-temperature phase diagram shows at least three different vortex-pinning regimes in the solid-vortex lattice region.

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