Correlated vortex pinning in Si-nanoparticle doped MgB$_2$

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The magnetoresistivity and critical current density of well characterized Si-nanoparticle doped and undoped Cu-sheathed MgB$_2$ tapes have been measured at temperatures $T \geq 28$ K in magnetic fields $B \leq 0.9$ T. The irreversibility line $B_{irr}(T)$ for doped tape shows a stepwise variation with a kink around 0.3 T. Such $B_{irr}(T)$ variation is typical for high-temperature superconductors with columnar defects (a kink occurs near the matching field $B_m$) and is very different from a smooth $B_{irr}(T)$ variation in undoped MgB$_2$ samples. The microstructure studies of nanoparticle doped MgB$_2$ samples show uniformly dispersed nanoprecipitates, which probably act as a correlated disorder. The observed difference between the field variations of the critical current density and pinning force density of the doped and undoped tape supports the above findings.

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INTRODUCTION

The discovery of superconductivity in MgB$_2$ compound$^1$ has aroused a great deal of interest in the scientific community.$^2$ Compared to high-temperature superconductors (HTS), MgB$_2$ has a lower transition temperature $T_c \approx 39$ K, but its simple composition, abundance of constituents and the absence of weak intergranular links$^3$$^4$$^5$ make the MgB$_2$ a promising material for applications at $T \geq 20$ K, which is above $T_c$s of conventional superconductors (LTS). Indeed, the simple preparation and rather high critical currents $J_c$ of composite MgB$_2$ tapes and wires$^6$$^7$$^8$$^9$$^{10}$ lend strong support to these expectations. Unfortunately, compared to practical LTS (NbTi, Nb$_3$Sn), MgB$_2$ exhibits weak flux-pinning$^{2}$,$^{11}$ which results in strong field dependence of $J_c$ and a low irreversibility field $B_{irr}(4.2$ K) $\approx 8$ T.$^2$

Several techniques, such as alloying$^{12}$,$^{13}$,$^{14}$,$^{15}$,$^{16}$,$^{17}$ particle irradiation$^{14}$,$^{15}$,$^{16}$,$^{17}$ and mechanical processing$^{18}$,$^{19}$ have been employed in order to improve the flux-pinning in MgB$_2$, but with limited success. In particular, proton irradiation$^{14}$ increased $B_{irr}$ at 20 K, but also suppressed low-field $J_c$, whereas alloying seems to enhance $J_c$, but has little effect on $B_{irr}$.$^{12}$,$^{13}$ Better results were recently obtained by adding nanoparticles to MgB$_2$.$^{19}$,$^{20}$,$^{21}$ It appears that a variety of nanoparticles considerably enhance the flux-pinning in MgB$_2$ over a wide temperature range $T \leq 30$ K. In particular, the addition of 10 wt% of SiC nanoparticles$^{20}$ yielded $B_{irr}(4.2$ K) $\approx 12$ T, which is higher than that of optimized NbTi$^{22}$.$^{23}$ The actual mechanism of the flux-pinning enhancement upon nanoparticle doping of MgB$_2$ is not well understood at present.

Here we present the results for magnetoresistance $R(T, B)$ and critical current $I_c(T, B)$ of MgB$_2$ tape doped with Si-nanoparticles, which reveal the flux-pinning mechanism associated with nanoparticle doping. In particular, $B_{irr}(T)$ of doped sample shows a kink at $B_{irr} \approx 0.3$ T, which is the signature of vortex pinning at correlated defects$^{22}$, whereas no kink is observed in undoped sample. The variation of critical current and pinning force density $F_p = J_c B$ with the field and temperature also show different pinning mechanisms in doped and undoped MgB$_2$, respectively.

EXPERIMENTAL PROCEDURES

Cu-sheathed MgB$_2$ tapes were prepared by in-situ powder-in-tube method$^8$. In the doped tape, in addition to Mg and B, 5 wt% of Si-nanoparticles with an average size $\sim 50$ nm was added. A low sintering temperature (670–690°C) and a short sintering time (several minutes) were employed$^{21}$ in order to avoid diffusion of Cu into the MgB$_2$ core$^{24}$. This resulted in rather porous, low density ($\sim 50\%$) cores. The core cross-sections were elliptic with areas 4.95·10$^{-3}$ and 4.8·10$^{-3}$ cm$^2$ for the doped and undoped tape, respectively. The sample lengths were approximately 1.5 cm and the voltage and current leads were soldered on Cu-sheathing. The magnetoresistance was measured with low-frequency ac method$^8$,$^{16}$ for $T \geq 28$ K in magnetic field $B \leq 0.9$ T perpendicular to a broad face of the tape and the current direction. $I_{RMS} = 1$ mA was used and the voltage resolution was 0.3 nV. Current criticals were measured on samples used in $R(T, B)$ measurements with the pulse method (sawtooth pulse with duration less than 10 ms and peak current of 200 A$^8$).

RESULTS AND DISCUSSION

The variation of the resistance with temperature ($28 \leq T \leq 300$ K) for our undoped and Si-doped tape (Fig. 1)
are typical for Cu-clad MgB$_2$ wires [24], with a larger resistance of the doped sample due to a larger distance between its voltage contacts.

Fig. 2 compares the superconducting transitions in fields $B \leq 0.9$ T for undoped and doped sample. As in other composite superconductors [25], the shape of these transitions is affected by Cu-sheathing. However, the onset of resistance (hence $T_c(R \to 0) = T_{c0}$) is not affected by sheathing [25], and the zero-field $T_{c0} = 38.2$ K for the undoped tape (Fig. 2a) is typical for bulk MgB$_2$ samples [2, 5, 24]. A strong shift of its $T_{c0}$ with magnetic field (i.e. $T_{\text{irr}}(B)$) reflects a weak flux-pinning in the undoped MgB$_2$. For the doped sample (Fig. 2b), zero-field $T_{c0} = 36.4$ K is lower than that of the undoped one, but the shift of its $T_{c0}$ with field is considerably smaller, which indicates an enhancement of flux-pinning (the expansion of the vortex-solid regime). Furthermore, values of $T_{c0}$ for the doped sample in $B \lesssim 0.3$ T are compressed within a rather narrow temperature interval, whereas those for the undoped one are more evenly spread throughout the explored field range.

Fig. 3 compares the irreversibility fields $B_{\text{irr}}(T)$ (defined by using the low-resistivity criterion $\rho_c = 5$ nΩcm) for our samples. For the undoped tape, both the magnitude and temperature variation of $B_{\text{irr}}$ are the same as the literature data for MgB$_2$ samples [2, 3, 16, 24, 26]. In particular, our values of $B_{\text{irr}}(T)$ are equal to those obtained from the onset of the third harmonic in the low-frequency ac susceptibility of a dense MgB$_2$ sample [20]. Approximately linear, $B_{\text{irr}}(T)$ variation for $T \leq 36$ K extrapolates to $B_{\text{irr}}(4.2$ K) $\approx 8.4$ T, which is a typical value for bulk MgB$_2$ [2].

The $B_{\text{irr}}(T)$ variation for the doped tape is very different from that of the undoped one (Fig. 3). Here, $B_{\text{irr}}$ increases rapidly with decreasing temperature down to 35.5 K, and shows slower, linear variation for $T \leq 35$
K. Such a stepwise $B_{irr}(T)$ variation is specific for HTS containing columnar defects \cite{23,27,28,29}, where the crossover in $B_{irr}(T)$ occurs around the matching field $B_0$, which is the field at which the vortex and columnar defect density $n_\phi$ are equal ($B_\phi = n_\phi \Phi_0$, $\Phi_0$ being the flux quantum \cite{23,29}). This crossover occurs because the pinning of interstitial vortices for $B_{irr} > B_\phi$ is less weak than for vortices residing onto the columns for $B_{irr} < B_\phi$.

From our crossover field $B_c \approx B_{irr}(35.5 \, \text{K}) \approx 0.3 \, \text{T}$ we estimate $n_\phi \approx 1.4 \cdot 10^{14} \, \text{m}^{-2}$, and the average distance between defects $\sim 80 \, \text{nm}$. The microstructural studies of the nanoparticle doped MgB$_2$ \cite{19,20,21} show finely dispersed precipitates within the MgB$_2$ matrix with sizes $\sim 10 \, \text{nm}$. For Si and SiC doped MgB$_2$ \cite{20,21} these precipitates are mainly Mg$_2$Si phase, and their average spacing is comparable to that estimated above. Therefore in our tape Mg$_2$Si nanoprecipitates, resulting from the reaction of Si-nanoparticles and Mg during the sintering, act analogously to columnar defects in HTS. This outcome appears rather surprising considering different nature and geometries of precipitates and columns, as well as the different nature of vortices \cite{23,30} in these materials. However, the matching effects are common in type-II superconductors \cite{31} and are not specific only to HTS.

A linear variation of $B_{irr}(T)$ for $T \leq 35 \, \text{K}$ in the doped sample extrapolates to $B_{irr}(4.2 \, \text{K}) \approx 11.5 \, \text{T}$, which is consistent with the other results for nanoparticle-doped MgB$_2$ \cite{19,20,21}, and is higher than $B_{irr}(4.2 \, \text{K})$ for NbTi. However, for $T > 33 \, \text{K}$, $B_{irr}$ of the doped sample is lower than that for the undoped sample, which is entirely due to its lower zero-field $T_c$. Indeed, a plot of $B_{irr}$ vs. reduced temperature $t_{irr} = T_{irr}(B)/T_{irr}(0)$ for both samples (inset to Fig. 3) shows that for all values of $t_{irr}$, $B_{irr}$ of the doped sample is higher than that of the undoped one. Therefore, vortex pinning in nanoparticle doped MgB$_2$ is enhanced with respect to that in undoped MgB$_2$ at all reduced temperatures.

Different vortex pinning mechanisms in our tapes imply also different field variations of their $J_c$ and $F_p = J_c B$. Fig. 4 compares the $J_c(B)$ variations of our samples for $T \geq 33 \, \text{K}$. The undoped tape (Fig. 4a) shows approximately exponential $J_c(B)$ variation, which is typical for MgB$_2$ samples \cite{2,14,15,20}. At low temperatures (high $I_c$), large self-field $\mu_0H_s$ ($H_s \approx I_c/c$, where $c$ is the circumference of the core) makes $J_c(B < \mu_0H_s)$ nearly constant, whereas at elevated fields ($B \rightarrow B_{irr}$) $J_c$ rapidly decreases to zero. From the experimental $J_c(B,T)$ curves (Fig. 4a) we obtained $F_p(B,T)$ ones, from which we determined the fields $B_{max}(T)$ at which the volume pinning force density reaches its maximum value $F_{p,\text{max}} = J_cB_{max}$. The field $B_{max}$ is an important parameter of the vortex pinning within the vortex-solid phase. In particular, in the case of dominant vortex pinning mechanism, the ratio $B_{max}/B_{irr}$ may reveal this mechanism. For the undoped tape we found $B_{max}/B_{irr} \approx 0.21$, which is similar to that observed in Nb$_3$Sn \cite{32} and is consistent with a commonly accepted grain boundary pinning mechanism for a bulk MgB$_2$ \cite{3}. In spite of a probably common vortex pinning mechanism in both bulk MgB$_2$ and Nb$_3$Sn, vortex pinning in MgB$_2$ is apparently weaker (lower $B_{max}$ and $B_{irr}$) than that in Nb$_3$Sn. The probable reason for that are larger grains, clean and narrow grain boundaries, and quite a large coherence length \cite{34} in MgB$_2$.

The $J_c(B)$ variation of nanoparticle doped MgB$_2$ sample (Fig. 4b) is very different from that for the undoped tape (Fig. 4a). The S-shaped $J_c(B)$ curves of the doped tape are reminiscent of those observed in HTS films, tapes and crystals containing columnar defects \cite{23}. Further, for the same reduced temperature $t = T/T_c$, the decrease of $J_c$ with $B$ in the doped tape is considerably smaller than that for the undoped one. Accordingly, the fields $B_{max}(t)$ are enhanced with respect to those of the undoped tape, which shows that nanoparticle doping enhances vortex pinning throughout the vortex-solid regime \cite{23}. Furthermore, the enhancement of $B_{max}(t)$ in the doped tape is larger than that of
$B_{irr}(t)$, which results in $B_{\text{max}}(t)/B_{irr}(t) \approx 0.29$ for the doped tape. Such $B_{\text{max}}/B_{irr}$ ratio is unlikely to arise only from the grain boundary pinning [22] and was earlier observed for HTS tapes [23] with a modest density of columnar defects ($B_p \lesssim 0.2$ T). Therefore, we propose that $B_{\text{max}}/B_{irr} \approx 0.29$ arises from the competition of two pinning mechanisms (for example, a grain boundary pinning and a core pinning at nanoprecipitates) as was the case in HTS tapes. A detailed investigation of $J_c(B)$ curves for a number of temperatures extending over a broad temperature range (which requires $T > 200$ K) is necessary in order to solve this problem.

In spite of 50% porosity, our tapes have large self-field $J_c$ (Fig. 4), which increase rapidly with decreasing temperature ($J_c(t) \simeq J_c(0)(1-t)^\alpha$, with $\alpha \approx 1.5$). In particular, the observed $J_c(0.9T_c) \approx 40$ kA/cm$^2$ for both tapes extrapolate to $J_c(20$ K) $\approx 550$ kA/cm$^2$, the value which was confirmed by the magnetic measurements of $J_c(20$ K) [21]. Therefore, fully dense MgB$_2$ tapes are expected to reach $J_c(20$ K) $\sim 10^6$ A/cm$^2$, which is above $J_c(4.2$ K) for the best Bi2223/Ag tapes.

In summary, we have shown that a uniform dispersion of Mg$_2$Si nanoprecipitates (resulting from the addition of Si-nanoparticles to Mg and B powders [20, 21]) not only enhances the flux-pinning in MgB$_2$ samples, but also introduces an additional pinning mechanism. In particular, we observed a step-wise variation of $B_{irr}(T)$ in nano-Si doped MgB$_2$ tape with a kink around $B_p \simeq 0.3$ T, which is reminiscent of the vortex pinning at correlated disorder in HTS [22, 23, 24, 25]. We also observed a corresponding difference in the shapes of $J_c(B)$ and $F_p(B)$ curves for the doped and undoped tape respectively. Although our results were obtained for MgB$_2$ tape doped with Si nanoparticles only, we believe that the above conclusions hold also for other MgB$_2$ samples doped with different types of nanoparticles [14, 20, 21], providing that these nanoparticles form uniformly dispersed non-superconducting nanoprecipitates.

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