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The combined influence of connectivity and disorder on J_c and T_c performances in $\text{Mg}_x\text{B}_2+10 \text{ wt } \% \text{ SiC}$

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The influences of connectivity and disorder on the critical current density J_c are discussed to clarify the different mechanisms of $J_c(H)$ enhancement in different magnetic field ranges. Excess Mg in $\text{Mg}_x\text{B}_2+10 \text{ wt } \% \text{ SiC}$ composites effectively improves the connectivity, as evidenced by both the resistivity properties and the Raman scattering. The promising $J_c(H)$ of $\text{Mg}_{1.15}\text{B}_2+10 \text{ wt } \% \text{ SiC}$ is attributed to both the high connectivity and the improved irreversibility field, H_{irr} , which is in agreement with the Raman fitting analysis. Raman scattering measurements suggest a strengthened electron- E_{2g} coupling and weakened disorder with Mg addition. © 2009 American Institute of Physics. [doi:10.1063/1.3253757]

I. INTRODUCTION

Study of the flux pinning mechanism in MgB_2 superconductor is of fundamental interest both for understanding its supercurrent properties and for breaking through the technical obstacles that inhibit its practical applications. The depairing current density, J_d , is $\sim 8.7 \times 10^8 \text{ A/cm}^2$ for pure MgB_2 , as estimated from the Ginzburg–Landau formula $J_d = \Phi_0/[3/(\sqrt{3})\pi\mu_0\lambda^2(T)\xi(T)]$, where Φ_0 is the flux quantum, μ_0 the permeability of vacuum, λ the penetration depth, and ξ the coherence length.¹ The self-field critical current density, $J_c(0)$, in the best connected samples indicates the ultimate current-carrying potential in the superconductor, which has been reported as $3.5 \times 10^7 \text{ A/cm}^2$ at 4.2 K and $1.6 \times 10^8 \text{ A/cm}^2$ at 2 K in clean films made by hybrid physical-chemical vapor deposition.^{2,3} These values are about 4% and 20% of the J_d values. Compared with these values, the $J_c(0)$ values in polycrystalline MgB_2 bulks and wires are very low and have great potential to be improved. Although the $J_c(H)$ values can be improved through chemical doping, they are still much lower than J_d and drop exponentially with applied magnetic field (H_A).⁴ How to keep the $J_c(H)$ close to J_d is the key to obtaining higher practicable supercurrents in MgB_2 . Chen *et al.*⁵ reported the possibility of improved J_c in low magnetic fields from a high Mg content in MgB_2 . Zeng *et al.*⁶ showed that 10 wt % Mg excess could improve the J_c values of MgB_2 wires over the entire range of measurement fields, 0–8.5 T.

In this work, the effects of excess Mg combined with nano-SiC codoping on the current transport properties and flux pinning mechanisms of MgB_2/Fe wires were studied systematically. To confirm the cooperation between the connectivity and lattice distortion, Raman scattering measurements were employed in this study. Raman scattering can

disclose the crystal strain caused by lattice distortion and chemical pressure, which is related to the flux pinning force (F_p).⁷

II. EXPERIMENTAL

The powder-in-tube process was employed to make practical MgB_2 wires from starting powders of Mg (99%), B (99%, amorphous), and SiC (<30 nm). The starting powders were weighed according to the molecular formulae for MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2+10 \text{ wt } \% \text{ SiC}$, with $x=1.00$, 1.15, 1.20, 1.25, and 1.30. Sintering was carried out at 650 °C for half an hour under a flowing argon gas atmosphere. The MgB_2 cores extracted from the annealed wires were ground and subjected to x-ray diffraction (XRD) measurements. The microstructures were observed by field emission gun scanning electron microscopy (FEG-SEM: JSM-6700F). The magnetic J_c was derived from the height of the magnetization loop ΔM using the Bean model: $J_c = 15\Delta M/[\pi a^3 h]$, where a and h are the radius and height of a cylindrical sample. The temperature dependent resistivity, $\rho(H, T)$, was measured with H applied perpendicular to the current direction, using the four-probe method in the temperature range from 15 to 300 K and the field range from 0 to 8.7 T. The upper critical field, H_{c2} , and the irreversibility field, H_{irr} , were defined as the magnetic field values at 90% and 10% of the superconducting transition on the $\rho(H, T)$ curve, respectively. The Raman scattering was measured with a confocal laser Raman spectrometer (Renishaw inVia plus) with a 100× microscope. The 514.5 nm line of an Ar⁺ laser was used for excitation.

III. RESULTS

The Mg excess and nanosize SiC doping effects on the phases and superconducting properties were first studied to explore their different functions in the behavior of the critical temperature, T_c , and the critical current density, J_c . Figure 1 shows the XRD patterns for the wire cores of MgB_2 ,

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FIG. 1. (Color online) XRD patterns of the MgB_2 cores in the iron clad wires of MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x=1.00, 1.15, 1.20, 1.25$, and 1.30) sintered at 650°C for 30 min.

$\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$. These XRD patterns reveal that nearly single-phase MgB_2 was obtained in pure, stoichiometric MgB_2 samples along with a small amount of MgO . A small amount of residual Mg could be found in $\text{Mg}_{1.15}\text{B}_2$, while the Mg_2Si content was obvious in $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$. Figure 2 shows the field dependence of J_c at 5 and 20 K in fields of 0–8.5 T. The pure MgB_2 shows a gradual decrease in J_c with increased measurement field at 5 and 20 K. The J_c values of $\text{Mg}_{1.15}\text{B}_2$ in low magnetic field, $< \sim 2.5 \text{ T}$ at 5 K and $< \sim 1.1 \text{ T}$ at 20 K, are the highest. Although this behavior cannot be observed directly at 5 K because the J_c values at 5 K are difficult to calculate due to the flux jumping, it is obvious that the J_c values for $\text{Mg}_{1.15}\text{B}_2$ wires at 20 K exceed those of all the other samples. The nano-SiC addition has improved the J_c over that of the pure stoichiometric MgB_2 in high measurement fields, $> \sim 7 \text{ T}$ at 5 K and $> \sim 4 \text{ T}$ at 20 K. However, the J_c values are very low at low field, as in previous results.⁴ The superconducting transitions are shown in the inset of Fig. 2. The additional Mg degrades the superconductivity of MgB_2 , and the T_c value of $\text{Mg}_{1.15}\text{B}_2$ is 0.7 K lower than that of the stoichiometric MgB_2 because the residual Mg is a source of impurity scattering effects. The T_c value, 35.3 K, for $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ is even lower than that of $\text{Mg}_{1.15}\text{B}_2$ because of both the C substitution (estimated as $\sim 2.2\%$, as shown in Table I⁸) and the impurity scattering.

FIG. 2. (Color online) A double-logarithmic plot of $J_c(H)$ estimated from the Bean model for MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x=1.00, 1.15, 1.20, 1.25$, and 1.30) wires at 5 and 20 K. The magnetic transformation behaviors are shown in the inset.

To take advantage of the doping effects of Mg and SiC , $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ samples were studied with $x=1.15, 1.20, 1.25$, and 1.30 . The residual Mg contents are similar in all the samples, as indicated by the XRD patterns in Fig. 1. As estimated from the XRD patterns, the C substitution content is $\sim 2.3\%$ in $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$, which is similar to that in $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$, and then it drops to 2.1% in $\text{Mg}_{1.20}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ and 1.8% in both $\text{Mg}_{1.25}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ and $\text{Mg}_{1.30}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$. It seems that the MgB_2 lattice excludes C atoms due to the excess Mg , as in the trend shown in Table I. The most impressive advantage of the Mg excess samples is the improved $J_c(H)$ in low magnetic field, especially at 5 K. However, the J_c values drop quickly for $\text{Mg}_{1.20}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$, $\text{Mg}_{1.25}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$, and $\text{Mg}_{1.30}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ because of the high impurity content. The decreased T_c values with increasing Mg content are responsible for the J_c drop at 20 K. The J_c performance of $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ improves over the whole range of applied magnetic field due to the optimum ratio of the component elements.

Although the T_c values decrease with the dopant contents, the different superconducting transition processes suggest quality differences between the MgB_2 crystals according to the normalized ac susceptibility, as shown in the inset of Fig. 2. The transition width, ΔT_c , is defined as the temperature difference between the onset value of the transition (the point where the ac susceptibility deviates from zero), $T_{c,\text{onset}}$, and the termination point of the transition (the point where the ac susceptibility reaches -1), $T_{c,\text{ter}}$. All the values of $T_{c,\text{onset}}$ and ΔT_c are listed in Table I. The superconducting transition of MgB_2 is quite narrow, with a ΔT_c of 0.98 K. However, it is even sharper for $\text{Mg}_{1.15}\text{B}_2$, with a ΔT_c of 0.85 K, which is attributed to the improved quality and connectivity of the MgB_2 because the flux is easy to expel with the decreased temperature. The transition of $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ is very broad, with a ΔT_c of 3.36 K, because of the strong disorder. Excess Mg in $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ is of benefit to the transition: the ΔT_c of $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ drops significantly to 1.69 K. More Mg addition decreases the value of ΔT_c to $\sim 1.55 \text{ K}$ in $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x > 1.20$). The stable ΔT_c values indicate that the quality cannot be improved with more Mg addition in $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$.

IV. DISCUSSION

A. Connectivity

Based on the collective pinning model,⁹ the disorder-induced spatial fluctuations in the vortex lattice can be clearly divided into different regimes according to the strength of the applied field: single-vortex, small-bundle, large-bundle, and charge-density-wave type relaxation of the vortex lattice. For high current purposes, the practicable J_c for MgB_2 is in the regions of single-vortex pinning and small-bundle pinning in the phase diagram. It is believed that the quality of the connectivity is responsible for the J_c performance in the single-vortex pinning regime (low magnetic field region) due to its weak field dependence, while the

TABLE I. Comparison of onset transition temperature ($T_{c,onset}$), transition width (ΔT_c), the lengths of the a -axis and c -axis parameters, the C substitution contents, the DOS $N(0)$, the resistivity values at 40 and 300 K, $\Delta\rho(300)$, and the active area fraction (A_F) for iron-clad wires of MgB_2 , $Mg_{1.15}B_2$, and Mg_xB_2+10 wt % SiC ($x=1.15, 1.20, 1.25$, and 1.30) sintered at 650 °C for 30 min.

Samples	MgB_2	$Mg_{1.15}B_2$	With 10 wt % SiC addition				
			MgB_2	$Mg_{1.15}B_2$	$Mg_{1.20}B_2$	$Mg_{1.25}B_2$	$Mg_{1.30}B_2$
$T_{c,onset}$ (K)	37.18	36.50	35.32	34.55	34.28	34.04	33.60
ΔT_c (K)	0.98	0.85	3.36	1.69	1.57	1.53	1.54
a -axis (Å)	3.0869	3.0881	3.0771	3.0766	3.0773	3.0786	3.0788
c -axis (Å)	3.5244	3.5238	3.5203	3.5201	3.5218	3.5224	3.5227
C content (%)	N/A	N/A	2.2	2.3	2.1	1.8	1.8
$N(0)$	0.354	0.354	0.332	0.331	0.333	0.336	0.336
$\rho_{40\text{ K}}$ ($\mu\Omega\text{ cm}$)	86.8	70.0	277.2	152.2	168.6	191.3	213.3
$\rho_{300\text{ K}}$ ($\mu\Omega\text{ cm}$)	171.6	125.5	422.5	246.3	269.8	307.7	347.3
$\Delta\rho(300)$ ($\mu\Omega\text{ cm}$)	84.8	55.5	145.3	94.1	101.2	116.4	134.0
A_F	0.106	0.162	0.062	0.096	0.089	0.077	0.067

disorder is responsible for the J_c performance in the small-bundle regime (high magnetic field) due to the strong F_p .

To explore the effect on connectivity of Mg excess, microstructures of all samples were observed with SEM, as

FIG. 3. SEM images of MgB_2 (a), $Mg_{1.15}B_2$ (b), MgB_2+10 wt % SiC (c), $Mg_{1.15}B_2+10$ wt % SiC (d), $Mg_{1.20}B_2+10$ wt % SiC (e), $Mg_{1.25}B_2+10$ wt % SiC (f), and $Mg_{1.30}B_2+10$ wt % SiC (g).

shown in Fig. 3. The grains in MgB_2 samples show an independent growth process, which is responsible for their isolated distribution. The grains in $Mg_{1.15}B_2$ have clearly melted into big clusters because the additional Mg can extend the liquid reaction time. The grain shapes in MgB_2+10 wt % SiC are different from that in pure, stoichiometric MgB_2 because the crystals are grown under strain due to the C substitution effect. The strain is also strong in $Mg_{1.15}B_2+10$ wt % SiC, as long bar grains can be observed under SEM. The strain is released in the high Mg content samples ($x>1.20$), judging from the homogeneous grain sizes and shapes. Compared with MgB_2+10 wt % SiC, the grain connectivity improved greatly with the increasing Mg addition. The grains are merged into big particles. This means that the grain boundaries have replaced the gaps between grains. However, more impurities are induced in forms such as residual Mg and MgO.

Compared with the microstructure observations, the resistivity is a more reliable and quantitative method to estimate the connectivity. The resistivity dependences on temperature are shown in Fig. 4. The $\rho_{300\text{ K}}$ and $\rho_{40\text{ K}}$ of MgB_2 wires are always high compared with thin films and bulks due to the low density of the wire samples. Although residual Mg exists in $Mg_{1.15}B_2$, its resistivity is decreased due to the

FIG. 4. Temperature dependence of resistivity curves of MgB_2 , $Mg_{1.15}B_2$, and Mg_xB_2+10 wt % SiC ($x=1.00, 1.15, 1.20, 1.25$, and 1.30).

highly connected grain boundaries. The resistivity increases greatly in $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ because of the strong band scattering effects due to the C substitution on B sites.¹⁰ The strong localization of the boron p_{xy} orbital on the boron plane, which is sensitive to the carbon substitution for boron, is responsible for the great resistivity enhancement. Additionally, the reduction in charge carriers has to be considered because the conducting holes are annihilated by the electrons introduced into the system by the substitutional carbon atoms. The most amazing effect of Mg excess is that the resistivity of $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ is reduced by $\sim 45\%$ with 15% Mg addition. This is because the grain boundary scattering effects are greatly weakened. Although a greater Mg excess ($x > 1.15$) ought to also reduce the resistivity of $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$, the increased scattering effects of the impurity phases make the resistivity a little higher than that of $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$.

A practical quantity to evaluate the connectivity is the active area fraction, $A_F = \Delta\rho_{\text{ideal}}/\Delta\rho(300 \text{ K})$, where $\Delta\rho_{\text{ideal}} = \rho_{\text{ideal}}(300 \text{ K}) - \rho_{\text{ideal}}(40 \text{ K}) \approx 9 \mu\Omega \text{ cm}$ is the resistivity of fully connected MgB_2 without any disorder,¹¹ and $\Delta\rho(300) = \rho(300 \text{ K}) - \rho(T_c)$. The A_F values for all the samples are listed in Table I. It should be noted that the connectivity is far removed from that found in ideal crystals, as reflected by the low A_F values. Although the A_F values of pure and 10% SiC doped MgB_2 are just 0.106 and 0.062, the additional Mg can improve them to 0.162 and 0.096 for 15 wt % Mg excess samples, respectively. High A_F values are the reflection of a broad channel of supercurrents, while impurities reduce the connectivity in large x samples, as shown in Table I. Considering the weak field dependence of J_c in the ranges of $H_A < \sim 0.7 \text{ T}$, 20 K and $H_A < \sim 2 \text{ T}$, 5 K, as shown in Fig. 2, it is believed that the high connectivity improves the supercurrent channels because the currents can easily meander through the well-connected grains.

B. H_{c2} , H_{c2} , and F_p

In contrast with the high connectivity mechanism in the single-vortex region, a strong flux pinning force is the dominant feature that keeps the J_c high in the small-bundle region. Most defects, lattice distortions, and grain boundaries are believed to act as flux pinning centers.^{7,12,13} Carbon substitution for boron in the MgB_2 lattice can cause great lattice distortion because of the shorter C–B bonds.¹⁰ Furthermore, H_{c2} and H_{irr} are significantly enhanced by the increased band scattering due to carbon substitution. The competitive J_c in high field for $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ and $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ should be ascribed to the enhanced H_{c2} and H_{irr} . Figure 5 compares the H_{c2} and H_{irr} performances for all the samples. The H_{c2} and H_{irr} values of pure, stoichiometric MgB_2 increase gently as the temperature is decreased, while the enhancement is a little slower in $\text{Mg}_{1.15}\text{B}_2$. $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ has high H_{c2} and H_{irr} values in the range of $T < \sim 24 \text{ K}$, and the additional Mg in $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ does not degrade the good performance of H_{c2} and H_{irr} . The high H_{c2} and H_{irr} of $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ are attributed to both the carbon substitution and the high amount of impurity phase.

FIG. 5. (Color online) Comparisons of H_{c2} (a) and H_{irr} (b) of MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x = 1.00, 1.15, 1.20, 1.25$, and 1.30).

The H_{irr} is responsible for the J_c performance in high magnetic field because the field dependence of F_p is proportional to $h^n(1-h)^m$, with $h = H/H_{irr}$, and the values of n and m depend on the different flux pinning mechanisms.¹⁴ The h dependence of $F_p/F_{p,\text{max}}$ is shown in Fig. 6. The x -axis is not normalized because the values of H_{irr} are higher than those in Refs. 15 and 16 due to the different standards for H_{irr} . The most striking phenomena in Fig. 6 are (i) the $F_p/F_{p,\text{max}}$ for $\text{Mg}_{1.15}\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ is effective both in the single-vortex region and in the small-bundle region; (ii) the $F_p/F_{p,\text{max}}$ for $\text{MgB}_2 + 10 \text{ wt } \% \text{ SiC}$ is the most effective in the small-bundle region; (iii) the $F_p/F_{p,\text{max}}$ values for MgB_2 and $\text{Mg}_{1.15}\text{B}_2$ are uncompetitive in the small-bundle region; and (iv) the $F_p/F_{p,\text{max}}$ values for $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x = 1.20, 1.25$, and 1.30) are also effective in the small-bundle region. These phenomena are in agreement with the J_c behavior shown in Fig. 2: (i) the J_c performance for $\text{Mg}_{1.15}\text{B}_2$

FIG. 6. (Color online) Comparisons of the $h(h = H/H_{irr})$ dependence of $F_p/F_{p,\text{max}}$ for MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_x\text{B}_2 + 10 \text{ wt } \% \text{ SiC}$ ($x = 1.00, 1.15, 1.20, 1.25$, and 1.30).

FIG. 7. (Color online) Ambient Raman spectra of MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, and $\text{Mg}_{1.30}\text{B}_2+10 \text{ wt } \% \text{ SiC}$ ($x=1.00, 1.15, 1.20, 1.25$, and 1.30) fitted with three peaks: ω_1 , ω_2 , and ω_3 . The dashed line indicates the vibration of the E_{2g} mode (ω_2) with different components.

+10 wt % SiC is promising over the whole range of the H_A ; (ii) the J_c values for $\text{MgB}_2+10 \text{ wt } \% \text{ SiC}$ drop slowly with the H_A , and it can be imagined that the J_c values are higher than those of $\text{Mg}_{1.15}\text{B}_2+10 \text{ wt } \% \text{ SiC}$ in the stronger H_A ; (iii) the $J_c(0)$ values of MgB_2 and $\text{Mg}_{1.15}\text{B}_2$ are outstanding, however, they drop quickly with H_A ; and (iv) the high impurity content degrades the J_c performance in low H_A for $\text{Mg}_x\text{B}_2+10 \text{ wt } \% \text{ SiC}$ ($x=1.20, 1.25$, and 1.30), whereas these impurities are also efficient flux pinning centers in high H_A . The high J_c of $\text{Mg}_{1.15}\text{B}_2+10 \text{ wt } \% \text{ SiC}$ is ascribed to the cooperation of the high A_F , high H_{c2} , and high H_{irr} . The H_{c2} and H_{irr} values of $\text{Mg}_x\text{B}_2+10 \text{ wt } \% \text{ SiC}$ ($x \geq 1.20$) are lower than those of $\text{Mg}_{1.15}\text{B}_2+10 \text{ wt } \% \text{ SiC}$, which is attributed to the high impurity content of the samples.

C. Raman spectra

Raman scattering is employed to study the combined influence of connectivity and lattice distortion. Chemical substitution¹³ and lattice distortion¹⁷ are expected to modify the phonon spectrum, by changing the phonon frequency and the electron-phonon interaction. The effects of C substitution include an increase in impurity scattering and band filling, which reduces the density of states (DOS) and alters the shape of the Fermi surface. The E_{2g} phonon peak shifts to the higher energy side, and the peak is narrowed with increasing x in $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$.¹³ As a carbon source, nano-SiC shows a similar influence, due to its C atoms, on the J_c , H_{irr} , H_{c2} , and even Raman spectra in MgB_2 . Figure 7 shows the Raman spectra fitted with three peaks: ω_1 , ω_2 , and ω_3 . The ω_1 and

FIG. 8. Fitted parameters of Raman shifts for ω_2 (a) and ω_3 (b), and FWHMs for ω_2 (c) and ω_3 (d). The sample labels are defined as A for $\text{Mg}_{1.15}\text{B}_2$, B for MgB_2 , C for $\text{MgB}_2+10 \text{ wt } \% \text{ SiC}$, D for $\text{Mg}_{1.15}\text{B}_2+10 \text{ wt } \% \text{ SiC}$, E for $\text{Mg}_{1.20}\text{B}_2+10 \text{ wt } \% \text{ SiC}$, F for $\text{Mg}_{1.25}\text{B}_2+10 \text{ wt } \% \text{ SiC}$, and G for $\text{Mg}_{1.30}\text{B}_2+10 \text{ wt } \% \text{ SiC}$.

ω_3 peaks are understood to arise from sampling of the phonon density of states (PDOS) due to disorder, while ω_2 is associated with the E_{2g} mode, which is the only Raman active mode for MgB_2 .¹⁸ A reasonable explanation for the appearance of ω_1 and ω_3 is the violation of Raman selection rules induced by disorder. All three peaks are broad, as in previous results, due to the strong electron-phonon coupling. The influence of ω_1 on the superconducting performance is negligible compared with those of ω_2 and ω_3 because of its weak contribution to the Raman spectrum. The frequency and full width at half maximum (FWHM) of ω_2 and ω_3 are shown in Fig. 8. Both ω_2 and ω_3 are hardened with SiC addition. The ω_2 frequency is reduced with further Mg addition, whereas the ω_3 frequency remains almost stable. The frequencies of ω_2 for the $x \geq 1.20$ samples are even lower than that of the pure, stoichiometric MgB_2 . The FWHM of ω_2 decreases with SiC doping, while the Mg excess remedies this trend. On the contrary, the ω_3 FWHM increases with SiC addition and becomes narrow with more addition of Mg.

The Raman scattering properties are the direct reflection of the phonon behaviors of MgB_2 . The parameters of Raman spectra vary with the composition of MgB_2 crystals and the influences of their surroundings, which depend on both the connectivity and the disorder of the samples. Furthermore, the disorder should be considered as intrinsic and extrinsic parts based on their different sources. The crystallinity and chemical substitution are believed to be responsible for the intrinsic disorder effects, while the grain boundaries and impurities are treated as responsible for the extrinsic disorder effects. The influences of intrinsic disorder on the basic characteristics of Raman spectra are significant because the

physical properties of MgB_2 depend on the intrinsic disorder. The Raman parameters can also be tuned by the extrinsic disorder. Especially in the samples with good connectivity, the influences of grain boundaries and impurities on the Raman spectra are taken into account because of their strain effects on the MgB_2 crystals.¹⁹ The differences between shifts and FWHMs in Raman spectra for MgB_2 , $\text{Mg}_{1.15}\text{B}_2$, MgB_2+10 wt % SiC, and $\text{Mg}_{1.15}\text{B}_2+10$ wt % SiC are mostly attributed to the intrinsic characteristics because of their different chemical compositions. The Raman spectra of Mg_xB_2+10 wt % SiC ($x>1.20$) can be considered as the gradual modification of that of $\text{Mg}_{1.15}\text{B}_2+10$ wt % SiC. The weakened C substitution effects are responsible for the decreased frequencies and slightly increased FWHMs of ω_2 with Mg addition. Accordingly, the FWHMs of ω_3 decrease due to the weakened lattice distortion. Although the A_F values are quite low for Mg_xB_2+10 wt % SiC ($x>1.20$), the effects of extrinsic disorder on Raman parameters are considerable through the $\text{MgB}_2\text{--MgB}_2$ and $\text{MgB}_2\text{--impurities}$ interfaces and the connectivity degrades with the increased x values due to the decreased numbers of $\text{MgB}_2\text{--MgB}_2$ interfaces. The high FWHM value of ω_2 is correlated with the high self-field J_c due to the high carrier density, while the high FWHM value of ω_3 is correlated with the high-field J_c because of the strong flux pinning force due to the large disorder. The FWHM behaviors show that high connectivity and strong disorder are best combined in $\text{Mg}_{1.15}\text{B}_2+10$ wt % SiC among all the samples.

D. Electron-phonon coupling

Not only are the Raman parameters related to the superconducting properties, but they are also related to the electron-phonon coupling (EPC) strength. The broad FWHM of ω_2 and the narrow FWHM of ω_3 in Raman spectra imply a strong electron- E_{2g} coupling in MgB_2 . According to the frequency and the linewidth of the E_{2g} mode reflected in the Raman spectra, direct evaluation of the contribution of the E_{2g} mode to the EPC is possible due to the negligible anharmonic effects in the system. The constant of the electron- E_{2g} coupling is given by the Allen equation:²⁰ $\Gamma_2 = 2\pi\lambda_{E_{2g}}N(0)\omega_2^2$, where $\lambda_{E_{2g}}$ is the strength of the electron- E_{2g} coupling and $N(0)$ is the DOS (per spin per unit energy per unit cell) on the Fermi surface and is the only electronic property explicitly occurring in this equation. The measured phonon frequency and phonon linewidth, in the absence of anharmonic contributions, are simply and directly related to the EPC constant, $\lambda_{E_{2g}}$. The total DOS at E_F in pure MgB_2 is taken as $N(0)=0.354$ states/eV/cell/spin, with the contribution from the σ band being 0.15 states/eV/cell/spin and that from the π band being 0.204 states/eV/cell/spin, respectively.²¹ The $N(0)$ is assumed to be constant for the small changes of electron and holes in $\text{Mg}_{1.15}\text{B}_2$ and MgB_2 . The factor of $N^\pi(0)$ dependence on the carbon concentration is about -1 in the range of $0\text{--}0.04$, while $N^\sigma(0)$ remains constant.²² Taking the C substitution contents shown in Table I into account, the $\lambda_{E_{2g}}$ values for the different samples can be obtained from the Allen equation, as shown in Fig. 9. These values are in agreement with 2.5 ± 1.1 ob-

FIG. 9. Calculated coupling constants of electron- E_{2g} , $\lambda_{E_{2g}}$, based on the Raman shifts and FWHMs of ω_2 according to the Allen equation. The sample labels are defined as A for $\text{Mg}_{1.15}\text{B}_2$, B for MgB_2 , C for MgB_2+10 wt % SiC, D for $\text{Mg}_{1.15}\text{B}_2+10$ wt % SiC, E for $\text{Mg}_{1.20}\text{B}_2+10$ wt % SiC, F for $\text{Mg}_{1.25}\text{B}_2+10$ wt % SiC, and G for $\text{Mg}_{1.30}\text{B}_2+10$ wt % SiC. The inset shows the calculated total EPC constants, λ_{EPC} , according to the McMillan formula.

tained for the $q=0.2\Gamma\text{--}A$ E_{2g} mode from inelastic x-ray scattering measurements.²³ It is unexpected that the $\lambda_{E_{2g}}$ of MgB_2+10 wt % SiC is just a little smaller than that of the pure, stoichiometric MgB_2 . A possible explanation is that both the samples are imperfect because of the low temperature sintering and impurity phases. The crystallinity is improved for all the Mg excess samples according to the higher $\lambda_{E_{2g}}$ values. The $\lambda_{E_{2g}}$ values in Mg_xB_2+10 wt % SiC ($x>1.20$) are even higher than that for $\text{Mg}_{1.15}\text{B}_2$, which is attributed to the low $N(0)$ values and the large amount of Mg excess. This phenomenon is in agreement with the strong FWHM values of ω_2 and the low FWHM values of ω_3 .

Although their E_{2g} mode contribution to the EPC is higher, the total EPC constants are degraded by the high impurity contents, which can be estimated with the McMillan formula,²⁴ as modified by Allen and Dynes:²⁵ $T_c = \langle\omega_{\log}\rangle / 1.2 \exp(-1.04(1+\lambda)/\lambda - \mu^*(1+0.62\lambda))$, where $\langle\omega_{\log}\rangle = (390 \times \omega_{E_{2g}}^2 \times 690)$ is the averaged phonon frequency,²⁶ with 690 and 390 cm^{-1} being the phonon frequencies of the other modes in the MgB_2 system (taken from Ref. 27), μ^* is the Coulomb pseudopotential, taken as equal to 0.13,²⁸ and λ is the EPC constant. Taking these values, the λ for each sample is calculated and is shown in the inset of Fig. 9. The λ of MgB_2 is the highest one because of its high T_c and low impurity content. Both Mg and SiC addition depress λ , which is in agreement with the C substitution effect and the high impurity contents shown in the XRD patterns.

V. CONCLUSIONS

In conclusion, connectivity and disorder are both important factors in improvement of the J_c performance of MgB_2 . The connectivity is responsible for the high J_c performance in the single-vortex regime, while strong disorder is responsible for the promising J_c in the small-bundle pinning regime. The promising J_c values in $\text{Mg}_{1.15}\text{B}_2+10$ wt % SiC are the result of optimized connectivity and disorder, which are reflected in the Raman spectrum, with both a strong E_{2g} mode and a strong PDOS. The Raman scattering measure-

ments imply that excess Mg is effective in improvement of the connectivity of MgB_2 grains, while nano-SiC is responsible for the great lattice distortion in the SiC-doped samples. The superconductivity transition is advanced in Mg excess samples according to the decreased ΔT_c . The EPC analysis shows that the excess Mg can also improve the electron- E_{2g} coupling. However, the impurity phases depress the total EPC strengths.

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