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High, magnetic field independent critical currents in (Ba,K)Fe_2As_2 crystals

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Each discovery of a new high temperature superconductor drives the expectation that advanced engineering of materials defect structures will enable effective vortex pinning and high values of the electrical current density. Here, we demonstrate that single crystals of the iron-based superconductor $Ba_{0.6}K_{0.4}Fe_2As_2$ with $T_c = 37.5$ K can accommodate an unprecedented large concentration of strong-pinning defects in the form of discontinuous nm-sized nanorods with no degradation of the superconducting transition temperature. At a temperature of 5 K, we find a critical current density of 5 MA/cm^2 that is magnetic field independent in fields up to 7 T. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4731204]

The recently discovered iron-based superconductors[1–3] display materials characteristics that make them very appealing for potential applications. Their reasonably high values of the superconducting transition temperature T_c, very high upper critical magnetic fields, and generally low anisotropy enable operation in economically interesting temperature-field ranges. Furthermore, these materials can be processed into high-quality thin-films[4–6] and reports show that the detrimental effect of grain boundaries on the supercurrent flow is significantly less pronounced[7] in these materials than for example in $YBa_2Cu_3O_{7-\delta}$-based conductors.[8] It is, therefore, imperative to determine the highest intra-grain critical currents that these materials can support. The maximum electrical current density is ultimately determined by the vortex pinning defects that can be tailored into the material without degrading the electrical pathway. Correlated disorder in the form of columnar defects created by self-assembled nanoparticles or amorphous tracks induced by energetic particle irradiation is especially effective for the study of high-field pinning properties of vortices, noticeably in cuprate high-T_c superconductors.[9–11] Previous studies of heavy-ion irradiation[12–14] of crystals and self-assembly of $BaFeO_2$ nanorods in films[15,16] have reported increases of the critical current in Fe-based superconductors. Here, we add a new dimension to these studies, by demonstrating the extremely high sustainability of irradiation induced nanorods with virtually no degradation of the superconducting transition temperature in $Ba_{0.6}K_{0.4}Fe_2As_2$ with $T_c = 37.5$ K. Our results are in sharp contrast to electron-doped pnictide superconductors where T_c is reduced with irradiation-induced defects.[17] Our irradiation induced defects result in the retention of high critical currents at high magnetic fields and at temperatures close to T_c. In addition, we demonstrate a desirable reduction in the superconducting anisotropy via irradiation in these new iron-based superconductors, underlining their benefit to the application of superconducting light-weight generators, motors, and cryogen-free magnet coils and MRI systems.[18]

$Ba_{1-x}K_xFe_2As_2$ (Ref. 2) is a member of the 122-family of Fe-based superconductors derived from the antiferromagnetic parent compound $BaFe_2As_2$, in which superconductivity is induced by hole-doping via partial substitution of Ba with K. At optimal doping, $x \simeq 0.4$, T_c reaches ~38 K, the highest among the 122-materials. A series of plate-like $Ba_{0.6}K_{0.4}Fe_2As_2$ crystals was irradiated at the ATLAS facility at Argonne National Laboratory with 1.4-GeV Pb-ions to fluences N corresponding to dose-matching fields of $B_\Phi = 4$, 6, 10, 21, and 50 T along the crystal c-axis; $B_\Phi = N\Phi_0$, where $\Phi_0 = 2.07 \times 10^{-15}$ Tm^2 is the flux quantum. Z-contrast images obtained in STEM (scanning transmission electron microscopy)[19] of the sample irradiated to a fluence of $B_\Phi = 21$ T (10^{12} ions/cm^2) are presented in Fig. 1. These images reveal that the irradiation produces segmented amorphous tracks with average diameter of 3.7 nm. The plan-view (Fig. 1(a)) shows the radiation tracks as a random array of black spots. The observed average track density amounts to ~35% of the value expected from the nominal irradiation dose. This shortfall is consistent with the discontinuous, segmented structure of the tracks. In the medium-resolution cross-section view in Fig. 1(b) and in the atomic-resolution image in Fig. 1(c) taken along the [100] direction, the tracks appear as discontinuous black streaks. Segmented tracks and a smaller than expected defect count in TEM-images were also observed in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ following irradiation with 200-MeV Au-ions.[12] The atomic-resolution image (inset of Fig. 1(a)) taken along the [001] direction shows Ba-As atomic columns and two dark spots associated with irradiation tracks. There is no crystal structure discernable inside the larger track indicative of an amorphous core. In contrast, atomic columns are visible inside the smaller track near the left edge of the inset. This observation is consistent with the segmented track structure, which results in the appearance of the crystal lattice projected from

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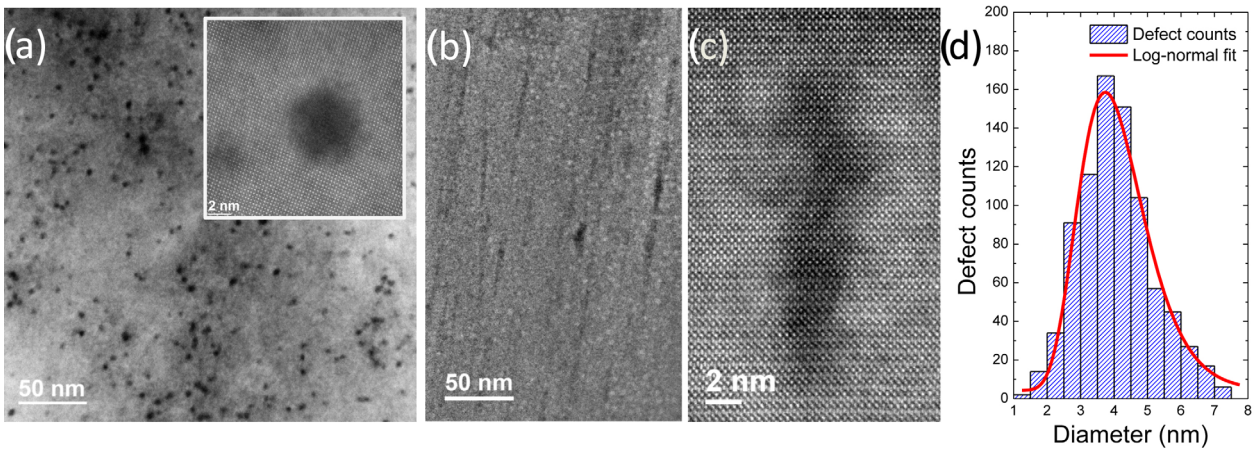

FIG. 1. Scanning transmission electron microscopy of the damage tracks. (a) Plan-view along the [001] direction revealing the damage tracks as black dots. The average areal coverage is ~5%. The inset shows an atomic-resolution image revealing the Ba-As columns and two tracks. The featureless black contrast of the larger track indicates amorphous material. (b) Cross-section view along the [100] direction. The tracks appear as discontinuous black streaks. (c) Atomic-resolution cross-section view revealing the layered crystal structure and a track segment in black. The white contrast in (c) and in the inset of (a) is indicative of strain fields surrounding the amorphous tracks. (d) log-normal distribution of the track diameters with average diameter of 3.7 nm.

the undamaged material above and/or below the defect track. The distribution of track sizes is well described by a log-normal distribution with average diameter of 3.7 nm (Fig. 1(d)). This size is considerably smaller than 6–8 nm typically observed in heavy-ion irradiated cuprate high-T_c superconductors[20,21] and consistent with the observation that tracks with radius of less than ~1.8 nm tend to be discontinuous.[21]

Fig. 2 summarizes the evolution of the superconducting transition of $Ba_{0.6}K_{0.4}Fe_2As_2$ with increasing irradiation dose as seen in magnetization and specific heat data. The temperature dependence of the magnetization (Fig. 2(a)) measured in a field of 10 G ∥ c after zero-field cooling reveals that the transition temperature and transition width do not change for doses up to 21 T. At $B_\Phi = 50$ T, T_c is suppressed by ~2 K and the transition broadens significantly. In comparison, the suppression of T_c of $YBa_2Cu_3O_{7-\delta}$ due to similar Pb-ion irradiation is considerably larger.[22] Since the amorphous cores in $Ba_{0.6}K_{0.4}Fe_2As_2$ are comparably small, one can expect that the damage to the surrounding material is also reduced, implying reduced suppression of T_c. The track morphology depends on parameters of the target material such as thermal conductivity, electronic structure, and electron-phonon coupling,[20,23] and the defect formation varies as the ions pass through the target material and lose their energy resulting in inhomogeneous defect structures. Nevertheless, the irradiation conditions introduced here allow the creation of defects at high concentrations without degrading T_c, a prerequisite for engineering high-field superconductors.

Fig. 2(b) shows the specific heat at the superconducting transition of the pristine and irradiated sample in zero field and in a field of 8 T parallel to the *c*-axis and *ab*-planes, respectively, giving information on the evolution of the intrinsic thermodynamic parameters. These data were obtained using a steady-state ac-calorimetric technique.[19] After irradiation to $B_\Phi = 21$ T, the height of the specific heat anomaly is clearly reduced, and the zero-field and in-field transitions broaden. The onset of the transition is nearly unchanged consistent with the magnetization measurements. The pristine sample displays a superconducting transition of height $\Delta C/T_c \sim 150$ mJ/mol K^2 at $T_c = 37.3$ K and upper critical field slopes of −6.7 T/K and −17 T/K along the *c*- and *ab*-directions, respectively, in good agreement with previous reports.[24,25] The corresponding values following irradiation are $T_c \simeq 37.0$ K, $\Delta C/T_c \simeq 120$ mJ/molK^2, and critical field slopes of −7.8 T/K and −16.7 T/K. We find a nearly 20% reduction in the superconducting anisotropy, Γ, which decreases from 2.55 to 2.15, a highly desirable feature for applications. The reduction of $\Delta C/T_c$ by ~20% is larger than expected on the basis of the areal fraction of amorphous tracks of ~5% as seen in electron-microscopy (Fig. 1(a)). This discrepancy indicates that material around the tracks is sufficiently damaged to strongly suppress its T_c perhaps out to twice the track radius, consistent with the extent of the white contrast seen in TEM. In contrast to the magnetization data, the specific heat data display a rather strong effect of irradiation to $B_\Phi = 21$ T, since the specific heat represents a true volume average whereas magnetization and resistivity are determined by the best superconducting path, that is, the remaining un-damaged material between the tracks.

The heavy-ion irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ displays a remarkable retention of high J_c values in high magnetic fields and at high temperatures as summarized in Fig. 3. Fig. 3(a) shows the critical current density J_c as determined from the magnetization hysteresis[19] in magnetic fields along the *c*-axis at 5 K and several irradiation doses. We parameterize the field dependence of the critical current density by the accommodation field B^* below which J_c is essentially field independent and the exponent α, which describes the rapid suppression of J_c in magnetic fields that are larger than B^* according to $J_c \sim H^{-\alpha}$. For $YBa_2Cu_3O_{7-\delta}$-coated conductors, typical values of B^* and α are 0.1-0.5 T and 0.4–0.7, respectively.[26–28] The extraordinary feature of our results is that after irradiation, J_c of $Ba_{0.6}K_{0.4}Fe_2As_2$ remains essentially field-independent at high levels of ~5 MA/cm^2 up to accommodation fields that exceed 5 T for the B_Φ=21 T sample. Although a direct comparison may be difficult due to self-field effects,[19] J_c of irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ decreases by only a factor of 1.3 when increasing the field to 7 T, whereas J_c of $YBa_2Cu_3O_{7-\delta}$-coated conductors decreases by a factor of 5-10 over the same

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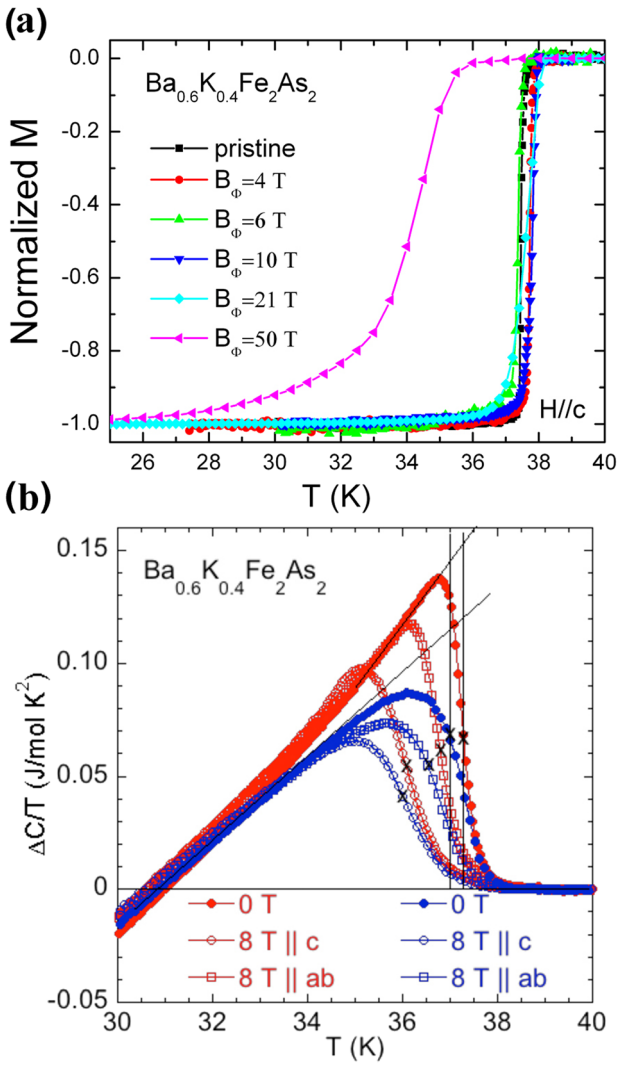

FIG. 2. (a) Temperature dependence of the magnetization, normalized to its low-temperature value, of a pristine crystal and crystals irradiated to various dose matching fields B_Φ measured on warming in a field of 10 G ∥ c after zero-field cooling. (b) Superconducting contribution to the specific heat ΔC/T at the transition in zero-field and in fields of 8 T along the c-axis and along the ab-planes for the pristine sample (in red) and for the irradiated sample with $B_\Phi = 21$ T (in blue). The crosses mark the locations of T_c defined through an entropy conserving construction as indicated by the solid lines for the zero-field data.

field range.[26,28,29] Thus, even though the zero-field critical current density of state-of-the-art $YBa_2Cu_3O_{7-\delta}$-coated conductors is higher than of our irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ crystals, owing to their exceptional in-field behavior, J_c-values at 4.2 K and in a field of 7 T∥c are in fact similar to those of $YBa_2Cu_3O_{7-\delta}$-coated conductors.

The large increase in B^* was not reported in earlier studies of J_c-enhancements in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ irradiated with 200-MeV Au-ions[12] and with 2.6-GeV U-ions.[13] The value of B^* of irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ increases in a systematic though sub-linear fashion with irradiation dose (Fig. 3(b)) up to ~5.4 T after irradiation to $B_\Phi = 21$ T. Work on pinning by random arrays of irradiation tracks in cuprate high-T_c superconductors has shown[9–11] that the roll-off or (at high temperatures) a maximum followed by a roll-off of J_c occurs near a field B^*, which is of the order of *(1/3–1/2)* B_Φ. Fig. 3(b) reveals that for small irradiation doses, B^* follows a linear $B_\Phi/3$—dependence reasonably well. The sub-linear behavior at higher B_Φ could originate from the finite probability that an incident Pb-ion hits a columnar track created by a previous ion.[10,30] Due to this effect, the number of (independent) tracks N_t (per sample area) does not increase linearly with ion-fluence but is instead given as[30] $N_t \simeq 1/\pi D^2\,(1 - \exp(-\pi D^2 B_\Phi/\Phi_0))$, where D is the track diameter, amounting to a correction of ~20% at a dose-matching field of 21 T. Such multiple hits can also partially account for the lower than expected defect count in TEM images.

At 5 K, the pristine sample has a low-field $J_c \sim 2$ MA/cm^2 which in fields larger than 0.3 T decays like $H^{-0.5}$ indicative of pinning by sparse strong pinning sites.[31,32] The low-field J_c undergoes a modest increase by a factor of 2–3 due to irradiation and shows no systematic dependence on the irradiation dose. Such dose-independence may be expected when all vortices are strongly pinned and a further increase in pinning sites will not increase J_c. The essentially field-independent J_c is the quintessential signature of this single-vortex strong-pinning regime. The de-pairing current density, the highest current superconducting electrons can carry before Cooper pairs break up, is given in Ginzburg-Landau theory by $J_d = \Phi_0/3\sqrt{3}\pi\mu_0\lambda^2\xi \sim 200$ MA/cm^2, using values of $\xi = 1.15$ nm for the coherence length and $\lambda = 175$ nm for the penetration depth as obtained from the analysis of the data in Fig. 2(b). *Albeit* rough, this estimate indicates that there is significant potential headroom for increasing J_c.

The gaps in the irradiation tracks have two opposing effects on vortex pinning: they are sources for expanding vortex loops (see situation (I) in the inset of Fig. 3(b)) limiting the critical current density of our present samples and, at the same time, they provide pinning of vortex kinks thus enhancing J_c and suppressing vortex creep (see situation (II) in the inset of Fig. 3(b)). In the single-vortex regime, the critical current density associated with expanding vortex loops can be estimated as[19] $J_c \approx (\sqrt{2}c\Phi_0/8\pi^2\lambda^2 L_G\Gamma)\left(\ln(L_G/\xi)\ln(D/2\xi)\right)^{1/2} \sim 1\times10^7$ A/cm^2 using a gap size of $L_G = 50$ nm (see supplemental information). When the expanding vortex loop becomes trapped on a neighboring track, two kinks form that travel up and down along the track until they get stuck at the next gap. The critical current density due to this latter process is estimated as $J_c \approx (c\Phi_0/16\pi^2\lambda^2 a_\phi)\ln(D/2\xi) \sim 1.7\times10^7$ A/cm^2, where a_Φ is the average track spacing. These estimates are in reasonable agreement with experiment and suggest that optimizing the gap length and track diameter could enhance the critical current density.

Irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ also retains high J_c on increasing temperature as shown in Fig. 3(c). Whereas J_c of the pristine sample is rapidly reduced by a factor of ~40 on increasing the temperature from 4.2 K to 30 K ($T/T_c = 0.81$), this reduction is more gradual and amounts to only a factor of ~7 for the sample irradiated to dose of $B_\Phi = 21$ T maintaining a value of $J_c = 0.32$ MA/cm^2 in 5 T at 30 K. These observations are in line with theoretical expectations that vortex pinning by strong correlated defects such as heavy-ion tracks is thermally much more robust than pinning by point disorder.[33] In comparison, J_c of state-of-the-art $YBa_2Cu_3O_{7-\delta}$ coated conductors[27,28,34] decreases by a factor of ~20 when increasing the temperature form 4.2 K to $T/T_c \sim 0.82$ (77 K).

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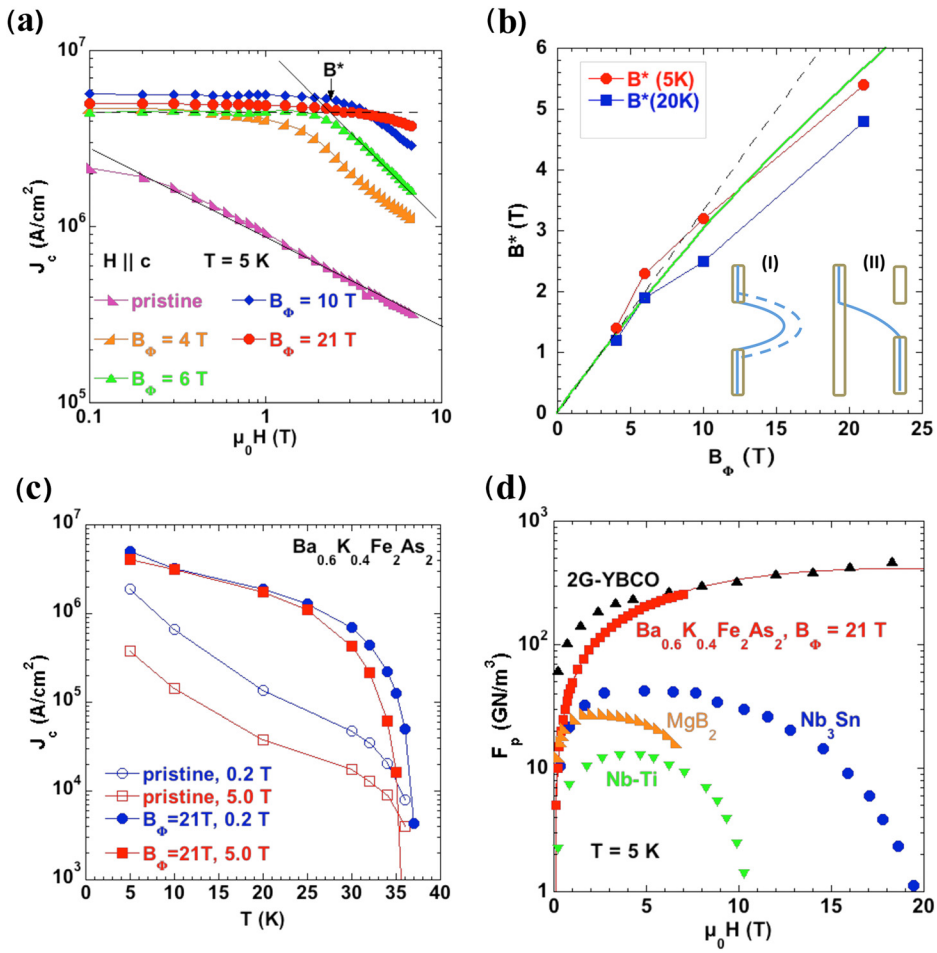

FIG. 3. (a) Field dependence of J_c at 5 K for various irradiation doses. Upon irradiation, J_c increases strongly and is field independent up to very high accommodation fields B*. The solid lines indicate field dependences of the forms $B^{-0.5}$ and B^{-1}, respectively; the dashed line is the baseline used for the determination of B* (arrow) for the B_Φ = 6 T—data. (b) Dependence of the accommodation field B* on irradiation dose B_Φ. The dashed line represents the relation $B^* = B_\Phi/3$. The solid green line represents the effect of multiple ions hitting the same track. The inset shows the formation of a vortex loop (I) and the pinning of a vortex kink (II) at a gap in the irradiation track. (c) Temperature dependence of J_c of the pristine and of the irradiated sample in low and high magnetic fields. (d) Comparison of the pinning force F_p at 5K of various superconducting materials. The red line is an extrapolation of the data of the B_Φ = 21T—$Ba_{0.6}K_{0.4}Fe_2As_2$ sample based on the scaling of the pinning force.[19] 2G-$YBa_2Cu_3O_{7-\delta}$ conductor,[29] NbTi,[36] Nb_3Sn,[37] MgB_2 (in H ∥ ab).[38]

The remarkable in-field performance of irradiated $Ba_{0.6}K_{0.4}Fe_2As_2$ is highlighted in Fig. 3(d) showing a comparison of the pinning force curve $F_p = \mu_0 H J_c$ of several high- and low-T_c materials. A comparison of the irreversibility fields may be found in Ref. 19. As seen in Figure 3(d), F_p of the $Ba_{0.6}K_{0.4}Fe_2As_2$ crystal is one order of magnitude higher than that of low-T_c materials and roughly a factor of ten smaller than that of state-of-the-art 2G-$YBa_2Cu_3O_{7-\delta}$ tapes at low fields, but become nearly equivalent at high fields.

In conclusion, we have shown that the irradiation of $Ba_{0.6}K_{0.4}Fe_2As_2$ with 1.4-GeV Pb-ions produces nanorods of small diameter of 3.7 nm. These irradiation conditions create an unprecedented concentration of strong pinning tracks while minimizing the degradation of the current-carrying material between the tracks. This pinning structure enables the remarkable retention of J_c in high-magnetic fields and at high temperatures that rivals those of current state-of-the-art 2 G YBCO coated conductors. Furthermore, even higher low-field J_c and high-field performance may be envisioned by optimizing the gap size between the nanorods or by incorporating mixed defect structures.[35]

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