Competition and cooperation of pinning by extrinsic point-like defects and intrinsic strong columnar defects in BaFe₂As₂ thin films

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We study the superconducting properties of Co-doped BaFe₂As₂ films on (La,Sr)(Al,Ta)O₃, as grown and after 3-MeV proton irradiations with doses up to 2×10^{16} cm⁻², as a function of temperature, magnetic field strength, and orientation by magnetization and transport. We study the pinning produced by the films' naturally grown strong correlated defects as well as the modified pinning landscape after the addition of point-like defects as a result of the irradiation. After irradiation, once the effect of a lower T_c is taken into account, the upper critical field (H_{c2}) remains unchanged, whereas the irreversibility field (H_{irr}) and the critical current density (J_c) decrease slightly at low fields. At high fields and low temperatures an overall increase in J_c is found, with J_c doubling along the *ab*-plane orientation due to a clear anisotropic contribution coming from the point-like defects induced by irradiation. We show that it is possible to achieve an "isotropic pinning" landscape at 9 T and 4 K when the two types of pinning centers are combined.

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I. INTRODUCTION

The so-called iron pnictide X122 family (XFe₂As₂, with X = Ca, Ba, Sr, or Eu) holds great promise as a model system to achieve a high critical current density J_c given the low anisotropy and high critical fields.¹⁻⁵ Although the detrimental effects of grain boundaries are less serious than in cuprates, they share the need for a biaxial texture to carry large amounts of currents.^{6–11} When Co-doped Ba122 films are grown by pulsed laser deposition on single-crystal substrates (or on single-crystal-like substrates), a high J_c , >1 MA cm⁻², can be obtained.^{12,13} A common feature in many of these films is that for a variety of substrates a large angular J_c peak is found centered around the c axis. This peak is the fingerprint of the presence of correlated defects^{13–15} and has been observed up to $\mu_0 H = 15$ T, indicating a very high density of these correlated defects.¹⁵ This has been confirmed by transmission electron microscopy (TEM) performed in Ba122 films on SrTiO₃-buffered (La,Sr)(Al,Ta)O₃ (LSAT) substrates, which show matching fields of $B_{\phi} \sim 8.5$ T.¹⁶ The *c*-axis J_c peak is so big that $J_c(||c|) > J_c(||ab|)$ in a very large range of fields and temperatures, in what is called "reversed anisotropy."^{14,15,17,18} This J_c angular peak is also very wide, reaching very close to the ab plane, an indication of very strong pinning.¹⁹ This angular behavior resembles that found in YBa2Cu3O7 (YBCO) films with self-assembled columns.^{17,18,20–23} A simple calculation of the pinning energy ϵ_n relative to the vortex line energy ϵ_l indicates that ϵ_n/ϵ_l in the Ba122 is at least 6 to 9 times higher than in the case of YBCO with self-assembled columnar defects.^{15,17,23}

Given the unprecedented strong pinning, this is a very good system to study the effects of extremely strong correlated defects, as well as to attempt to increase the overall pinning further. Also, this system lends itself to exploration of the effects of the controlled addition of different types of pinning centers, since it is totally dominated by correlated defects in a very large range of magnetic fields (**H**) and temperatures

(*T*). Having a complex pinning landscape has been shown to be beneficial, in particular, by reducing the negative effects of fast-flux relaxation at low fields.¹⁷ Although combining different types of defects has been pointed out to be one of the reasons for the high J_c in cuprates, there are theoretical results that predict that a J_c decrease due to competition can also occur. To evaluate whether a particular combination will be beneficial or detrimental, both types of defects have to be taken into account; for example, randomly dispersed point-like defects can disrupt the resulting glass phase of columnar defects (Bose glass)^{24,25} but not the smectic order due to insulating planes.^{26,27}

Combining dissimilar strong pinning centers to create a complex pinning landscape, with the prospect of increasing the overall pinning, has been a long-time goal in applications²⁸ and has been widely attempted by chemical methods.^{17,22,29-31} In several studies it has been observed that the initial pinning seems to decrease upon the addition of a second type of defect.^{17,22,29} Whether this occurs because of the competition between the pinnings to different defects or because the density and/or shape of the original defects are affected by the introduction of the new ones is still an open question. It has been speculated that one kind of pinning becomes less effective in the presence of another type, although definitive answers were not obtained.^{17,32} Samples with combinations of different types of defects to create a complex pinning landscape have been obtained by means of the introduction of two types of defects by irradiation,³² as well as by exploiting their natural occurrence.33

Given the correlated nature of the defects in Co-doped Ba122 films, the addition of randomly distributed point defects is a natural choice, both to increase the pinning and to study the interaction between point-like and correlated defects. In this paper we present the effect of the irradiation with 3-MeV protons up to doses of 2×10^{16} cm⁻². The 3-MeV protons are known to create from one to a few tens of atom displacements,³⁴ producing mainly random point defects

and also some nanoclusters of a few nanometers in size. In principle, the damage produced by 3-MeV protons should not affect the nonsuperconducting nature of the natural correlated defects in the films. One thing to point out is that this pinning landscape is different from that obtained with BaZrO₃ (BZO) addition in YBCO studied by Maiorov *et al.*¹⁷ since it is comprised of correlated and point-like defects, while in the case of Ref. 17 the pinning was mostly from correlated defects and nano-particles.

In this study we find that, once the small decrease in T_c is accounted for, H_{c2} remains unchanged after irradiation, whereas H_{irr} decreases slightly at low and intermediate fields. Also, J_c for $\mu_0 H < 1$ T decreases after irradiation. A clear improvement in the pinning properties is observed at higher fields, where an increase in J_c is found for all field orientations, with J_c doubling along the *ab* planes. The results of this study provide knowledge that will allow us to explore ways to increase J_c further, by engineering complex pinning landscapes exploiting the combination and minimizing the competition of different types of defects.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

Films were deposited by pulsed laser deposition on LSAT substrates^{9,12,35,36⁻} similar to that in Ref. 37. Upon further adjusting deposition conditions, highly homogeneous films were obtained with $T_c = 21.5$ K and $\Delta T_c \sim 1$ K. These films have an improved crystalline quality and drastically reduced Fe phases with respect to that found earlier.^{37,38} Cross-sectional TEM studies show a sharp interface, with the film chemically homogeneous through the thickness and very few Fe precipitates.¹⁵ Films with thicknesses (δ) of 0.2–0.5 μ m were patterned using dry etching into 5- to $20-\mu$ m-wide bridges. The same procedure was used to define a rectangular geometry in the films for magnetization measurements, with typical values of w = 1 mm and l = 1-3 mm, with w and *l* being the width and length of the film, respectively. After etching, a protective layer of gold was deposited on top of the films selected for magnetization measurements.

Transport measurements, both linear (resistivity, ρ) and nonlinear (V-I curves) were carried out in applied magnetic fields up to 9 T in the maximum Lorentz force configuration ($\mathbf{J} \perp \mathbf{H}$), with **H** applied at an angle Θ from the film's normal using a rotating probe in a variable temperature insert in a Quantum Design Physical Property Measurement System. Resistivity data $\rho(\Theta, T, H)$ were taken using a current density of 5 A cm⁻². V-I curves were analyzed using a 1 μ V/cm criterion to determine the critical current I_c .

The magnetization (*M*) measurements were performed using a superconducting quantum interference device (SQUID) magnetometer for **H** parallel to the *c* axis. The J_c values were calculated using the Bean critical state model,^{39,40} with $J_c = \frac{20\Delta M}{w(1-w/3l)}$, where ΔM is the difference in magnetization between the top and the bottom branches of the hysteresis loops.

Films were irradiated with 3-MeV protons simultaneously with single crystals in Ref. 41 at cumulative doses of 1×10^{16} cm⁻² (F1) and 2×10^{16} cm⁻² (F2).⁴² The average distance between the introduced defects as estimated using the SRIM code is 3.6 and 2.8 nm for F1 and F2, respectively.⁴¹



FIG. 1. (Color online) $\rho(T)/\rho(200 \text{ K})$ for a 0.4 μ m film, as grown (AG), first irradiation (dose, $1 \times 10^{16} \text{ cm}^{-2}$; F1), and second irradiation (accumulated dose, $2 \times 10^{16} \text{ cm}^{-2}$; F2). Inset: Transition temperature (T_c) vs irradiation dose (f). The slope of the decrease in T_c with irradiation dose f is $\partial T_c/\partial f \approx -0.5 \text{ K}/10^{16} \text{ cm}^{-2}$. Explicitly, $\partial T_c/\partial f = -0.52 \pm 0.01 \text{ K} \times 10^{-16} \text{ cm}^2$ or $-0.54 \pm 0.06 \text{ K} \times 10^{-16} \text{ cm}^2$ for T_c determined using the $0.9\rho_n$ or onset criterion, respectively, as shown in the figure.

III. RESULTS AND DISCUSSION

A. H_{c2} and H_{irr} phase diagrams

In Fig. 1 we show how from the $\rho(T)$ curves we determine the upper critical field and temperature H_{c2} and T_{c2} as well as the irreversibility line (H_{irr}, T_{irr}) using $0.9\rho_n$ and $0.01\rho_n$ criteria, respectively, with $\rho_n = \rho(25 \text{ K})$. As shown in Fig. 1, after each irradiation with 10^{16} cm^{-2} , T_c decreased by about 0.5 K, less than what we found in Ba(Fe_{0.925}Co_{0.075})₂As₂ single crystals, which showed a T_c drop of 0.7-1 K per 10^{16} cm^{-2} dose.⁴¹ A decrease in T_c near ~1.5 K/10¹⁶ cm⁻² was also found by Nakajima.⁴³ However, in the single-crystal cases T_c was near 25 K, almost 4 K higher than in the films shown here. The smaller decrease in T_c might be related to the higher degree of disorder initially present in the thin films.

In Fig. 2 we observe that $H_{c2}(T)$ shifts to a lower T for F1 and F2, for both $\mathbf{H} \parallel c$ and $\mathbf{H} \parallel ab$. Indeed, when plotted as a function of $t = T/T_c$, H_{c2} scales very well for both orientations. Similarly the shape of $H_{c2}(\Theta)$ remains unchanged when irradiated, as can be observed in Fig. 3. The lack of change in the shape of $H_{c2}(T,\Theta)$ is consistent with the small coherence length in these compounds, which is not affected by the introduction of disorder, and can be described in the framework of the so-called "Swiss cheese" model, which considers the local suppression of the superfluid density in the proximity of defects.^{44,45}

A different effect is observed for H_{irr} . As previously reported, the pinning in as grown (AG) films from the correlated pinning is so strong that at intermediate fields ($\mu_0 H \approx 1$ T), $H_{irr}(||c) > H_{irr}(||ab)$, the inverse of what is expected taking solely the effects of the anisotropy into account.¹⁵ The increase in H_{irr} from correlated defects arises from their ability to reduce the entropy of the vortex lattice, reducing the average vortex



FIG. 2. (Color online) H_{c2} as a function of temperature, T, for the **H** $\parallel c$ and **H** $\parallel ab$ planes. Inset: H_{c2} as a function of reduced temperature, $t = T/T_c$, for the same field orientations.

displacement $\langle |u| \rangle$.^{19,24} In Fig. 3, a small but clear decrease in $H_{\rm irr}$ is observed at $\mu_0 H < 3$ T upon irradiation. This is more pronounced at lower fields, where the effect of the correlated defects is more important, as indicated by the presence of a *c*-axis peak in H_{irr} . At $\mu_0 H = 1$ T [see Fig. 3(c)] it is clear that most of this decrease comes from washing out the c-axis peak in $H_{\rm irr}$. This indicates that the addition of randomly dispersed point defects is reducing the effectiveness of the correlated pinning to increase the H_{irr} . This negative effect on the signature of the Bose glass was predicted by Hwa, Nelson, and Vinokur.²⁵ In very clean YBCO crystals, where a first-order phase transition was observed, irradiation with protons produced a decrease in the vortex melting transition temperature.⁴⁶ The reasonable concern could be raised that the lower $H_{\rm irr}$ is due to a widening of the superconducting transition after irradiation. However, the transition width (ΔT_c) remains unchanged with irradiation within 0.01 K, measured as $\Delta T_c = T(0.90\rho) - T(0.01\rho)$. Also, the effect of a bigger ΔT_c should become more important at higher fields, rather than vanishing as in Fig. 3(a).

A couple of conclusions can be drawn from the results presented so far, namely, that proton irradiation does not affect H_{c2} besides the decrease in T_c ; also, the decrease in H_{irr} indicates a competition between randomly distributed point-like defects and columnar defects. The former prevents the reduction of the vortex entropy and concomitant increase in H_{irr} produced by columnar defects. This negative effect is strongest at magnetic fields where the correlated defects are more effective (~1 T) and becomes less or not important at higher fields (~9 T), at which correlated defects affect the H_{irr} much less.

B. Magnetic field and temperature dependence of J_c

We now turn our attention to the effects on the critical currents, starting with the temperature dependence. In Fig. 4 we show the J_c temperature dependence measured by magne-



FIG. 3. (Color online) T_{c2} and T_{irr} as a function of magnetic field orientation, Θ , for $\mu_0 H = 1, 3$, and 9 T.

tization [Fig. 4(a)] for $\mathbf{H} \parallel c$ and by transport [Fig. 4(b)] for $\mathbf{H} \parallel c$ and $\mathbf{H} \parallel ab$.

Once the effect of the lower T_c has been taken into account by using $J_c(t)$, we still find a small decrease in J_c at all *t* at low fields. The decrease in J_c at *self-field* (H = 0) is significant, from $J_c = 3.5$ MA cm⁻² down to close to 2.5 MA cm⁻². However, this J_c quickly levels off, and $J_c(t)$ at 1 T is the same for AG, F1, and F2 within the resolution of the magnetization measurements as shown in Fig. 4(a). The decrease in J_c is the opposite to what was found in Ba122 single crystals, where increases in J_c were observed.^{41,43} However, an important difference in the effects on the vortex pinning



FIG. 4. (Color online) (a) $J_c(t)$ for as grown (AG), first irradiation (F1), and second irradiation (F2) extracted from magnetization measurements, with $t = T/T_c$. Inset: J_c vs $1 - t^2$ for AG films with thickness $\delta = 0.2$ and 0.4 μ m for $\mathbf{H} \parallel c = 0.01$ T. (b) J_c vs $1 - t^2$ at $\mu_0 H = 1$ T for F2 with $\mathbf{H} \parallel c$ and $\mathbf{H} \parallel ab$ measured by transport using a 1 μ V/cm criterion. (c) Fitting of the J_c data for $\mathbf{H} \parallel c$ at 1 T for different expressions.

properties is expected from those observed in Ba122 single crystals, since these films have a J_c of several MA cm⁻² to start with, while Ba122 single crystals have an initial J_c well under 1 MA cm⁻².^{41,43,47} Thus, we do not expect an increase in J_c as great as seen in the crystals. Indeed, we observe no significant enhancement of J_c but, rather, a decrease for the magnetization measurements (**H** $\parallel c$) shown in Fig. 4. We note that the $J_c(sf)$ values at 4 K are still slightly lower than those

reported by Nakajima *et al.*^{48,49} after irradiation with heavy ions but higher than that reported by the same authors with protons.⁵⁰ It is possible that lower proton doses are needed for maximizing J_c at lower fields.

Similar trends are also observed in cuprate single crystals and films; while YBCO crystals show very important enhancements in J_c under proton (or heavy-ion) irradiation, thin YBCO films with a much higher J_c to start with show only an incremental J_c enhancement or even a J_c decrease at high T due to the lower T_c after irradiation.^{28,34,51}

When J_c is plotted as a function of $(1 - t^2)$ on a log-log scale a clear straight line is observed. For all samples and fields studied we find that the temperature dependence can be fitted with a $J_c \propto (1 - t^2)^n$ for almost the entire temperature range, with the results for *n* reported in Table I. In contrast, for YBCO high- J_c films a $(1 - t^2)^n$ is only observed at high temperatures, down to approximately $t \sim 0.45$ (T = 40 K).³³ In Fig. 4(b) we also observe that J_c is higher for **H** $\parallel c$ than for **H** $\parallel ab$ in the entire temperature range measured, indicating that the strong pinning from columnar defects always dominates.

In all the films we measured we find that $n \sim 2.8$ and that n remains unchanged after the irradiations. This n exponent is similar to the values obtained in Ba122 single crystals after proton irradiation.⁴¹ However, this value of n is puzzling because $n \sim 2.75$ is the expected exponent for the δl type of pinning,¹⁹ but the angular dependence clearly shows that the pinning for the AG films comes from columnar defects and thus corresponds to δT_c rather than δl .¹⁹ Fitting of $J_c(T)$ in early YBCO films indicated a δl pinning,⁵² however, those films had much smaller J_c values than the current ones.^{28,33} For state-of-the-art films and coated conductors, n = 1.5 is found, more consistent with a δT_c scenario.

A possible origin for variation in *n* could be attributed to different types of pairing mechanisms, since the effect of (non-magnetic) impurity scattering depends on the pairing of the carriers. Thus, an exotic type of pairing mechanism may lead to a dependence of the coherence length on impurity scattering.¹⁹ However, this argument is not applicable here since $n \sim 1.5$ is found for Co-doped Ba122 single crystals,⁴¹ and both films and crystals have the same pairing mechanism. We can speculate that correlated pinning might work differently, thus

TABLE I. Parameters *n* extracted from J_c vs $(1 - t^2)^n$ for two films of different thicknesses, AG ($\delta = 0.4 \,\mu$ m) and AG2 ($\delta = 0.2 \,\mu$ m), and at different magnetic fields for AG, F1, and F2.

Sample	Field (T)	n
AG	0.01	2.46
AG	0.3	2.73
AG	1.0	3.02
F1	0.01	2.56
F1	1.0	2.85
F2	0.01	2.45
F2	0.1	2.73
F2	0.3	2.75
F2	1	3.07
AG2	0.01	2.54
AG2	0.3	2.84
AG2	1	3.09

comparative studies between Ba122 films and YBCO films with self-assembled columnar defects are under way. Indeed, *n* is not field independent, growing as *H* increases, as reported in Table I. This could be associated with an effect of vortex interactions, since the exponents of δl and δT_c are calculated for a single-vortex scenario.¹⁹ Also, other mechanisms such as the renormalization of the pinning potential can be used to explain the J_c temperature dependence; depending on the nature of the defects [two-dimensional (2D), 1D, or pointlike], different temperature thermal smearing exponents are expected.¹⁹ This type of analysis has been successful in fitting $J_c(T)$ in high temperatures superconductors.^{53,54} According to Nelson and Vinokur, $J_c(T) \propto \exp(-3(T/T^*)^2)$ for $T_1 < T <$ T_{dp} , with T_1 being the temperature at which the entropy of flux-line wandering plays a significant role in determining the localization length, and $T^* = \sqrt{U_0/\gamma} b_0.^{24,55}$ When applied to fit the data shown in Fig. 4(c) we find that $J_c(T)$ can fit satisfactorily for $t \leq 0.6$ with $T^* = 0.95T_c$. Following Nelson and Vinokur for $T \ge T_{dp}$, we expect $J_c(T) \propto (T^*/T)^4$. As clearly shown in Fig. 4(c), $(T^*/T)^4$ fails to capture the temperature dependence of J_c at higher temperatures. It is worth noting that $(1 - t^2)^n$ captures J_c completely over the whole temperature range. Following Appendix D in Ref. 55 we find that $T_1/T_c = 0.985$; this is a direct result of the low value of Gi $\sim 10^{-5}$ for Ba122, in contrast to Gi $\sim 10^{-2}$ or $Gi \sim 10^{-1}$ for YBCO or $Bi_2Sr_2CaCu_2O_8$, respectively. This very high value of T_1 would indicate that columnar pinning is barely affected by thermal fluctuations.

The decrease in J_c at low fields is also clearly shown in Fig. 5, where we plot the $J_c(H)$ for $\mathbf{H} \parallel c$ at T = 15, 10, and 4.5 K for AG, F1, and F2, as well as T = 9.4 K for F2. Three clear field regimes can be observed, namely, a field-independent J_c at low fields (<0.1 T), a power-law regime ($J_c \propto H^{-\alpha}$) for $0.1 \leq \mu_0 H \leq 1$ T, and then a faster decrease in J_c at higher fields. It can be appreciated that J_c decreases upon irradiation with $J_c(AG) > J_c(F1) > J_c(F2)$. If



FIG. 5. (Color online) $J_c(\mathbf{H} \parallel c)$ for as grown (AG), first irradiation (F1), and second irradiation (F2) for 4.5, 10, and 15 K measured by magnetization; also shown, T = 9.4 K for F2. Also shown are the fits to $J_c \propto H^{-\alpha}$ with $\alpha \approx 0.3$.

the measurement is performed at equally reduced temperatures (e.g., T = 10 K for AG and T = 9.4 K for F2), a smaller reduction is found, consistent with the $J_c(t)$ shown in Fig. 4. As *H* increases, the differences among AG, F1, and F2 become smaller, with a slight crossover at higher fields ($\mu_0 H > 1$ T). In summary, J_c for **H** $\parallel c$ shows a small but clear decrease upon irradiation, especially at low fields, with a possible positive effect at higher fields ($\mu_0 H > 1$ T).

In the power-law regime $J_c \propto H^{-\alpha}$ values of $\alpha \sim 0.5$ have been widely reported in the YBCO family as well as in pnictides.^{41,47,56-58} However, for these films we observe a much smaller value of $\alpha \sim 0.3$ and thus a slower J_c decay with magnetic field. Such a reduction in α , and the consequent improvement in $J_c(H)$, has also been observed in YBCO films with correlated defects produced by different methods.^{17,18,23,59,60} This indicates a shared physics in the pinning of both YBCO and Ba122. Indeed, Co-doped Ba122 films grown on different substrates present strong correlated defects.^{13,15} The similarities are not complete, though, since in YBCO with correlated defects the value of α decreases with decreasing T, and at low temperatures the power law is lost.^{17,57}

In short, the temperature dependence of J_c over the whole temperature range measured can be fitted with a $(1 - t^2)^n$ with $n \sim 2.8$ and is not affected by proton irradiation. A similar trend is found in the field dependence of J_c , where $J_c \propto H^{-\alpha}$ with $\alpha \sim 0.3$ and remains unchanged after proton irradiation. Although J_c for **H** $\parallel c$ decreases at low fields with irradiation, a crossover occurs at higher fields around 1 T.

One would expect that if the crystalline quality of the sample is not greatly compromised, an enhancement or, in the worst case, a leveling of J_c should occur. Thus, the decrease in J_c at low fields can be taken as (a) a decrease in the crystalline quality of the sample or (b) a competition between the effect of random pinning and the already present columnar defects. If the former is the reason for the decrease in J_c , this should also affect the overall J_c . In contrast, if the latter is the case, we should observe an increase in J_c for other orientations.

C. Angular dependence of J_c

A more complete representation of the effects on the pinning properties can be obtained from the angular measurements. In Fig. 6 we plot $J_c(\Theta)$ at 1 T for AG, F1, and F2 at 10 K and for F2 at 9.4 K ($t = T/T_c = 0.45$). We observe a small decrease in J_c near the *c* axis, but no decrease in the value for **H** || *ab* despite the reduction in T_c . When AG and F2 are compared at the same *t* this translates into an improvement in J_c after irradiation for the *ab*-plane orientation. This is reasonable; randomly distributed point-like defects in an anisotropic superconductor have a $J_c(\Theta)$ contribution that is maximum along the *ab* plane and minimum in the *c* axis.^{58,61}

At lower temperatures we observe a similar effect, except that at T = 4 K (see Fig. 7) the increase around the *ab* planes is already evident when comparing $J_c(\Theta)$ at the same T, without the need for comparison at the same t. Even more, J_c at the c axis is barely down for F1.

Before we continue with the analysis of the effects of proton radiation, it is worth exploring the $J_c(\Theta)$ for AG. One important observation is that, different from what is found in cuprates with correlated defects, the *c*-axis peak is visible down



FIG. 6. $J_c(\Theta)$ for as grown (AG), first irradiation (F1), and second irradiation (F2) for 10 K and $\mu_0 H = 1$ T measured by transport. Also included are the measurements for T = 9.4 K for F2.

to the lowest temperatures measured (T = 2 K, $t \sim 0.09$). For YBCO samples with correlated defects (both films and crystals, with defects generated by heavy-ion irradiation, selfassembled columns, or twin boundaries) at similar reduced and absolute temperatures the *c*-axis peak is not observed.^{17,57,62} We also see that the angular dependence for AG at 9 T is dominated by correlated defects near the c axis, indicating the strong pinning and high density of the defects, with reversed J_c anisotropy. A second small peak near the *ab* planes is also observed. This peak is most likely due to the layered structure, however, the low anisotropy and relatively large size of the coherence length ξ_c on the c axis make it unlikeyly to be a smectic vortex solid as in YBCO at low temperatures.^{63,64} Nevertheless, the pinning landscape of these Co-doped Ba122's is dominated by columnar pinning throughout the entire temperature and field phase diagram.

The columnar-dominated pinning with $J_c(||c) \gg J_c(||ab)$ for AG becomes almost flat after the first dose of protons as shown in Fig. 7(c), with J_c at $\Theta = 90^{\circ}$ being almost twice for F1 as it was for AG. It is also clear by comparing Figs. 7(a)-7(c)that as H increases, the effect of the proton irradiation becomes more important, with J_c enhanced for all field orientations. For F2, J_c decreases with respect to F1 on the c axis. At 9 T, $J_c(\Theta)$ for F2 is leveled off near the c axis (but with a clearly visible *c*-axis peak) and shows the characteristic angular dependence coming from the presence of randomly dispersed point-like defects in an anisotropic superconductor, that is, a $J_c(\Theta)$ that is minimum for $\mathbf{H} \parallel c$ and maximum for $\mathbf{H} \parallel ab$.⁶¹ At this point it is not possible to perform a satisfactory anisotropic scaling analysis to study the dependence of the anisotropy of J_c in the T-H phase diagram. The positive effects along the ab plane are still increasing for F2, indicating that higher doses can still be applied before J_c is maximized at this orientation; a piece of information obtained only through the angular measurements. This results suggests the use of proton-induced defects (or point-like defects induced by other means) in applications where a high J_c along the *ab* plane is important.



FIG. 7. $J_c(\Theta)$ for as grown (AG), first irradiation (F1), and second irradiation (F2) for 4 K and $\mu_0 H = 1$, 6, and 9 T measured by transport.

It is also evident that a second structure develops near the *ab* planes, marked with an ellipse in Fig. 7(c) at $\Theta \sim$ 80°. This type of angular shoulder has been extensivly observed in cuprate high temperature superconductors with nanoparticles.^{17,22,65–67} This suggests the presence of nanoparticles in our films and points out to the need to learn more about the microstructure of AG films as well as the effects of proton irradiation. To that end, we are performing TEM studies to investigate the size and distribution of the defects. It has been shown that proton irradiation in YBCO produces about 70% point defects and 30% clusters.⁴⁶ However, it is not clear how the migration mechanisms that form the clusters work in a different and much more disordered matrix.

In short, the angular dependence measurements show that J_c is dominated by extremely strong columnar defects at all the fields (up to 9 T) and temperatures (down to 2 K) measured. For the AG films, the reversed anisotropy is present at all temperatures with $J_c(||c|) > J_c(||ab)$ due to the strong pinning of the correlated defects and low anisotropy. After irradiation we see a modest decrease in J_c for $\mathbf{H} \parallel c$ but a clear and robust increase in J_c at other orientations from an anisotropic random pinning contribution. This enhancement becomes more important at higher H, almost doubling J_c along the ab planes.

IV. SUMMARY AND CONCLUSIONS

We have investigated the superconducting properties of Codoped Ba122 films on LSAT, AG and after proton irradiations at doses up to 2×10^{16} cm⁻², as a function of temperature, magnetic field strength, and orientation by magnetization and transport. AG films show strong correlated defects that absolutely dominate the vortex pinning at all temperatures and fields, hence they are very appropriate for studying defect addition and for increasing the overall J_c .

The comparison of films AG and after proton irradiation shows that, once the effect of the lower T_c is taken into account, H_{c2} remains unchanged, whereas H_{irr} decreases slightly at low and intermediate fields, showing no variation at higher fields ($\mu_0 H \sim 9$ T). This indicates a negative effect from point-like defects on the pinning dominated by correlated defects.

After irradiation, J_c decreases at lower fields but the negative effects subside around 1 T. At high fields and low temperatures an overall increase in J_c is found, with

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 J_c doubling along the *ab*-plane orientation due to a clear anisotropic contribution coming from the point-like defects. We show that it is possible to achieve an "isotropic pinning" landscape at 9 T and 4 K when the two types of pinning centers are combined.

Our findings indicate that although an improvement in pinning can compensate the orientational pinning of correlated defects in the high-field and low-temperature region, also a clear decrease in the effectiveness of the correlated pinning is found in both H_{irr} and J_c at lower fields. This also points out that there is no "magic-bullet" approach when maximizing pinning and that careful consideration of the phase diagram region to be improved is needed. Detailed and controlled studies of the combination of different types of defects could lead to further insight into vortex pinning, revealing areas of cooperation and/or competition.

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