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Enhancement of critical current density and vortex activation energy in proton-irradiated Co-doped $BaFe_2As_2$

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The effect of proton irradiation in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystals is reported. We analyze temperature dependence of the current density and normalized flux relaxation rate in the framework of the collective-creep model. The glassy exponent and barrier height for flux creep are directly determined by Maley's method. Our model functions for barrier height and critical current density in the absence of flux creep are explained by the superposition of δT_c and δl pinnings. We also approach true critical current density by means of the generalized inversion scheme, and the obtained result is in reasonable agreement with our model function. The proton-irradiation effect on temperature dependence of the current density and normalized relaxation rate can be summarized as doubling of the barrier height at the beginning of flux creep.

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In high-temperature superconductors, many interesting phenomena in vortex dynamics are discovered, such as giant-flux creep and thermally activated flux flow, and the theories to describe them have been elaborated in past decades.[1] Especially, collective pinning with a weak-pinning potential by the quenched disorder and collective creep of vortex bundles give rise to intriguing experimental results, such as "plateau" observed in the temperature-dependent normalized relaxation rate ($S \equiv |d \ln M/d \ln t|$) (Ref. 2) in contrast to the linear increase with temperature predicted by the Anderson-Kim model in low-temperature superconductors. Recently discovered iron-based superconductors (IBSs) have relatively high critical temperatures (T_c) and large critical current densities (J_c). Besides, the magnetization hysteresis loop in this system is quite similar to that in Y-Ba-Cu-O, and magnetic relaxation measurements have revealed that IBSs also show giant-flux creep, which implies that IBSs and cuprate superconductors share common vortex physics. Moreover, how the introduction of the artificial pinning center affects flux dynamics or J_c is also interesting.[3,4] In Y-Ba-Cu-O, J_c is enhanced, and glassy behavior remains basically the same after proton (H^+) irradiation, which is known to introduce point defects. This is also expected in IBSs. In fact, Haberkorn *et al.* recently reported that H^+ irradiation does not affect the $H - T/T_c$ phase diagram in $Ba(Fe_{0.925}Co_{0.075})_2As_2$.[5] It is important to clarify how vortex dynamics is affected by H^+ irradiation in IBSs.

As we mentioned above, IBSs are suitable candidates to check whether glassy behavior of vortices is universal in all high-temperature superconductors. Since this system is twin free and less anisotropic, it enables us to discuss intrinsic pinning and dynamic properties of vortices without complications. However, it is difficult to synthesize large and clean crystals, especially, in LnFeAsO (Ln represents lanthanoid, the so-called "1111" system).[6] This prevents us from discussing vortex dynamics due to strong inhomogeneities unless we use local probes. Since high-quality single crystals are readily available in the so-called "122" crystals $AEFe_2As_2$ (AE represents alkaline earths),[7,8] it is also possible to discuss the details of vortex pinning in IBSs with global magnetic measurements. Actually, homogeneous flow of the superconducting current in this system has been confirmed by magneto-optical measurement.[9] This is why we choose the optimally Co-doped $BaFe_2As_2$ single crystal.

In this paper, we report the effect of proton (H^+) irradiation in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystals. We analyze it in the framework of collective-creep theory with temperature-dependent shielding currents J and normalized relaxation rates S. The glassy exponent and barrier height for flux creep are directly determined. Our model functions for barrier height and critical current density in the absence of flux creep are explained by the superposition of δT_c and δl pinnings. We also approach the true critical current density by means of the generalized inversion scheme, and the obtained result is in reasonable agreement with our model function. Effects of H^+ irradiation on $J(T)$ and $S(T)$ can be summarized as doubling of the barrier height at the beginning of flux creep.

Optimally Co-doped $BaFe_2As_2$ single crystals were grown by the FeAs/CoAs self-flux method. Fundamental properties of this system have been reported elsewhere.[9,10] All samples are cleaved to be thin plates with thicknesses less than ~ 20 μm. This value is much smaller than the projected range of 3-MeV H^+ for $Ba(Fe_{0.93}Co_{0.07})_2As_2$ of ~ 50 μm, calculated by the stopping and range of ions in matter.[11] The 3-MeV H^+ irradiation was performed parallel to the c axis at 40 K at the National Institute of Radiological Sciences, Heavy Ion Medical Accelerator in Chiba (NIRS-HIMAC). The total dose of the measured sample was 1.2×10^{16} cm^{-2}.[12] Magnetization was measured in a commercial superconducting quantum interference device magnetometer (MPMS-XL5, Quantum Design) with a magnetic field parallel to the c axis. The main features of vortex dynamics in the H^+-irradiated sample analyzed in this paper were briefly reported in Ref. 13. To clarify the effect of H^+ irradiation on the vortex system, we also measured a pristine sample similar to the "unirradiated" sample in Ref. 14. Current densities calculated by the Bean model are denoted

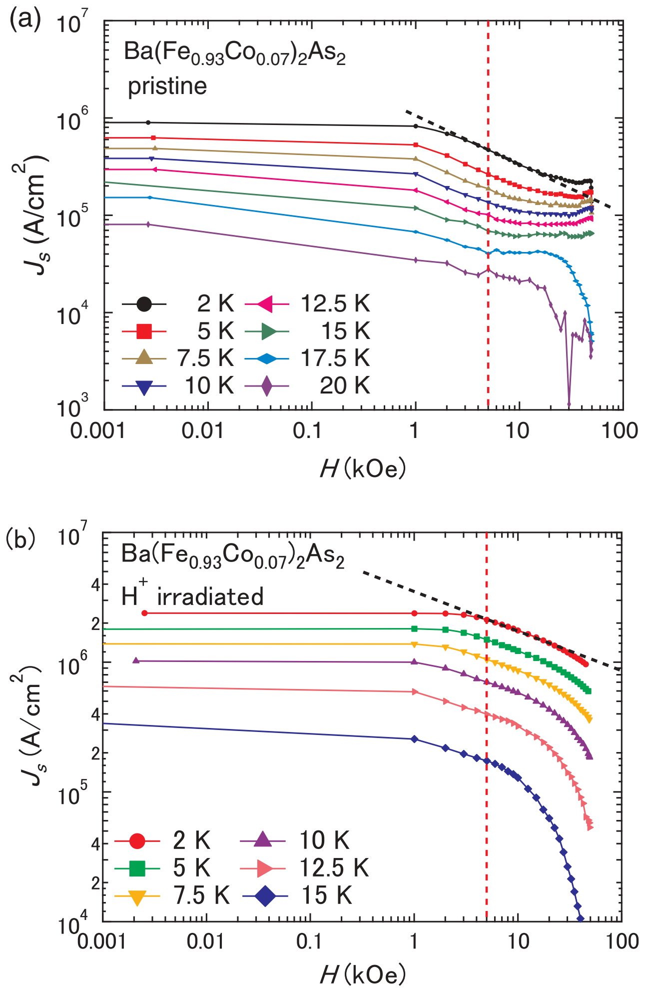

FIG. 1. (Color online) Field dependence of J_s in (a) pristine and (b) H$^+$-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ at several temperatures. The dotted line on ● (2 K) shows power-law decay of (a) $H^{-0.5}$ and (b) $H^{-0.3}$, respectively. The vertical line indicates the field where we discuss the vortex dynamics $H = 5$ kOe.

as J_s in field-sweep measurements (s is an abbreviation for sweep) or simply J in relaxation measurements.

Figure 1 shows the magnetic-field dependence of J_s in (a) pristine and (b) H$^+$-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$. It is obvious that H$^+$ irradiation enhances J_s from 1×10^6 to 2.5×10^6 A/cm^2 at 2 K under the zero field. In the pristine sample, J_s is nearly constant below 1 kOe, followed by power-law decay $H^{-\alpha}$ at a field of 2–10 kOe with $\alpha \sim 0.5$. As discussed by van der Beek and co-workers, these behaviors at low fields are attributed to sparse strong-point-pinning centers as in the case of Y-Ba-Cu-O films.[15,16] At a glance, it seems inappropriate to analyze it in the framework of collective-pinning-collective-creep and vortex glass theory. However, since the strong-point-pinning contribution for temperature (T) dependence of current density (J) is smaller than the weak-collective-pinning contribution (see Fig. 6 or 9 in Ref. 15), we can approximate $J(T)$ only by the contribution from collective creep (pinning). In the H$^+$-irradiated sample, it is basically the same as the pristine one, although there is a wide-crossover region with $\alpha \sim 0.3$ between the low-field plateau and the $H^{-0.5}$ region. Such a weak-field dependence has also been observed in YBa$_2$Cu$_3$O$_7$ and YBa$_2$Cu$_4$O$_8$ films by Griessen *et al.*, and they concluded that single vortex creep is achieved in this region.[17]

To elucidate the vortex dynamics, it is important to measure the (static) magnetic relaxation rate $S \equiv |d \ln M/d \ln t|$ in both samples, where M is magnetization and t is time from the moment when the critical state is prepared. In order to discuss temperature dependence of vortex dynamics, we have to fix the magnetic field. However, as we mentioned above, there is a strong-pinning background so that we have to select a field where field dependence of J_s is similar for all temperatures to exclude the field-dependent strong-point-pinning effect. Besides, we should carefully keep away from the fishtail effect with nonmonotonic field dependence of J_s at high fields and the self-field effect at low fields, which disturb the direct extraction of typical parameters for vortex dynamics. Based on these considerations, we select $H = 5$ kOe in both samples, shown as the vertical broken lines in Fig. 1. The insets of Fig. 2 show the temperature dependence of S. According to collective-pinning theory,[18] this is described

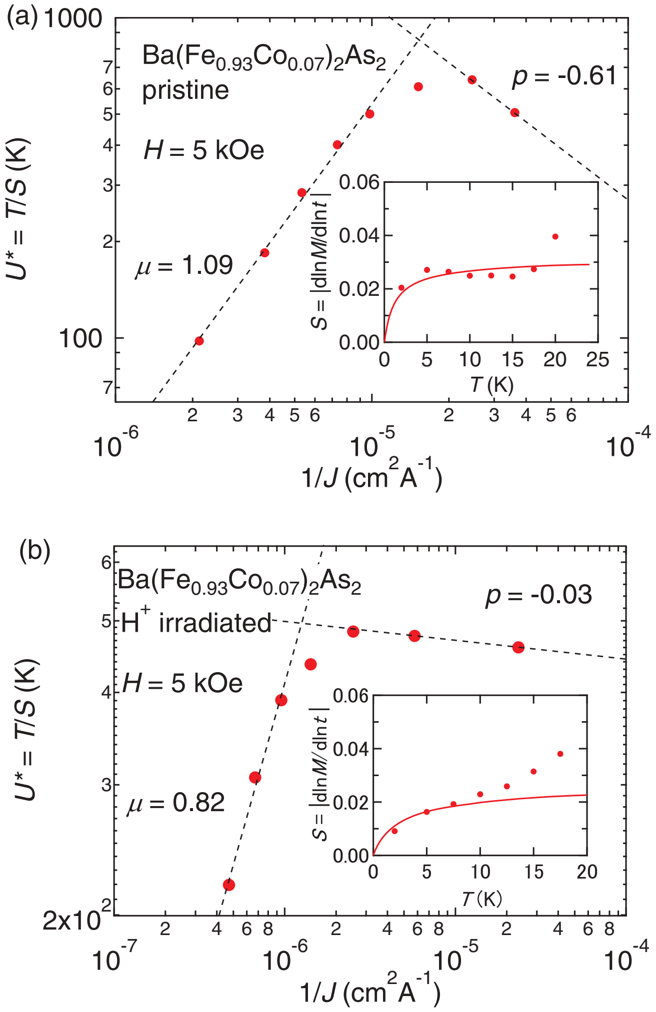

FIG. 2. (Color online) Inverse current-density dependence of effective pinning energy U^* in (a) pristine and (b) H$^+$-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$. Inset: Temperature dependence of normalized relaxation rate S. The solid line indicates fitting by Eq. (1).

as

$$S = \frac{T}{U_0 + \mu T\, \ln(t/t_{\mathrm{eff}})}, \tag{1}$$

where U_0 is the temperature-dependent flux activation energy in the absence of flux creep, $\mu > 0$ is a glassy exponent for elastic creep, and t_{eff} is the effective hopping attempt time. One of the most remarkable results extracted from this equation is the prediction of plateau in the intermediate temperature range if $U_0 \ll T$.[2] The value of plateau $S \sim 1/[\mu\, \ln(t/t_{\mathrm{eff}})]$ falls in the range of 0.02-0.04 theoretically, which has been confirmed in Y-Ba-Cu-O.[2] The inset of Fig. 2(a) is quantitatively consistent with this behavior as observed in other IBSs.[14,19,20] This proves the validity of applying collective-pinning theory to IBSs. Here, we emphasize that it is quite important to determine the value of μ in discussing vortex dynamics since μ includes information on the size of the vortex bundle in collective-pinning theory. In a three-dimensional system, it is predicted as $\mu = 1/7, 3/2, 7/9$ for single-vortex, small-bundle, and large-bundle regimes, respectively.[21] Inverse current-density dependence of effective pinning energy $U^* = T/S$ is convenient to extract this value.

We can define the inverse power-law form of flux activation energy $U(J)$ as

$$U(J) = \frac{U_0}{\mu}[(J_{c0}/J)^{\mu} - 1]. \tag{2}$$

Combining this with $U = T\, \ln(t/t_{\mathrm{eff}})$ extracted from the Arrhenius relation, we can deduce the so-called "interpolation formula,"

$$J(T,t) = \frac{J_{c0}}{[1 + (\mu T/U_0)\ln(t/t_{\mathrm{eff}})]^{1/\mu}}, \tag{3}$$

where J_{c0} is the temperature-dependent critical current density in the absence of flux creep. From Eqs. (2) and (3),

$$U^* = U_0 + \mu T\, \ln(t/t_{\mathrm{eff}}) = U_0\,(J_{c0}/J)^{\mu} \tag{4}$$

is derived. Thus, the slope in the double logarithmic plot of U^* vs $1/J$ gives the value of μ, shown in the main panels of Fig. 2. In this way, we evaluate $\mu = 1.09$ and 0.82 for pristine and H^+-irradiated samples, respectively. Note that $\mu \simeq 1$ in the pristine crystal is often reported in Y-Ba-Cu-O (Ref. 22) and IBSs.[23] Contrary to the above prediction of $\mu > 0$, a negative slope is observed at small J. This negative slope is often denoted as p in plastic-creep theory with $p = -0.5$ and is confirmed experimentally.[24] Our evaluation of $p = -0.61$ in the pristine sample is very similar to this value.

To determine the actual flux activation energy, we employ the extended Maley's method.[25] Since temperature dependence of U is not considered in the original Maley's method,[26] it is impossible to scale U in a wide range of J, even if the glassy exponent is unique. In order to solve this problem, appropriate temperature dependences of U_0 and J_{c0} are assumed as follows:

$$U_0(T) = U_{00}[1 - (T/T_c)^2]^n, \tag{5}$$

$$J_{c0}(T) = J_{c00}[1 - (T/T_c)^2]^n. \tag{6}$$

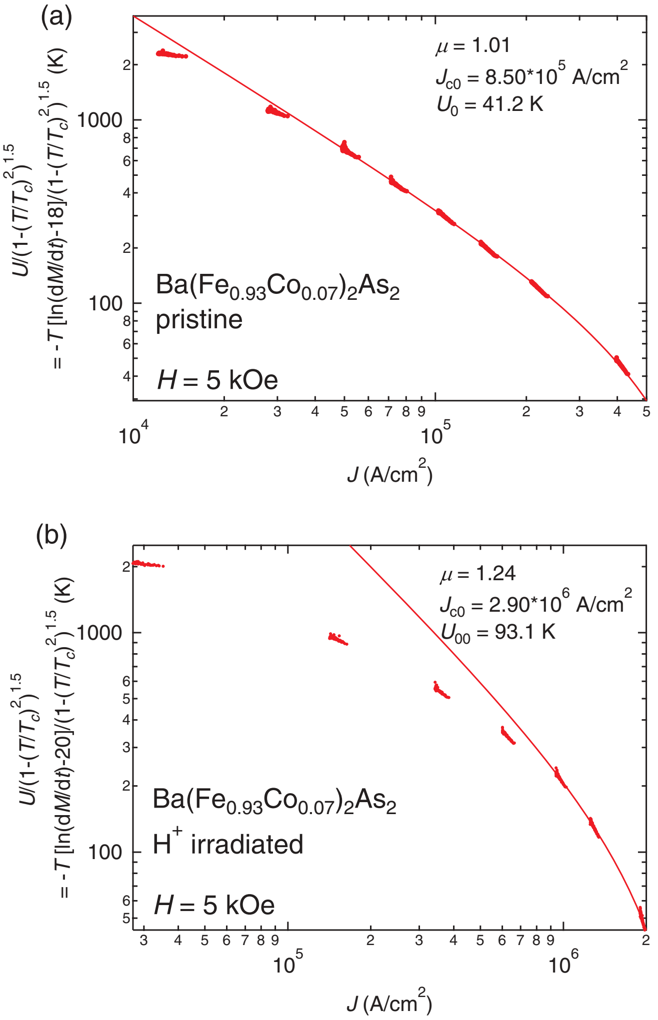

FIG. 3. (Color online) Current-density dependence of U in (a) pristine and (b) H^+-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ constructed by the extended Maley's method. The solid line indicates power-law fitting in the large J region.

In order to simplify the problem, we choose the same exponents in Eqs. (5) and (6). Here, exponent n is set to 3/2 as in the case of Refs. 22 and 25, whereas, $(1 - T/T_c)^{3/2}$ is selected in $Ba_{1-x}K_xFe_2As_2$.[23] $U = -T\, \ln[dM(t)/dt] + CT$, and $C = \ln(B\omega a/2\pi r)$ is assumed as a constant, where B is the magnetic induction, ω is the attempt frequency for vortex hopping, a is the hopping distance, and r is the sample radius. We select $C = 18$ and 20 for pristine and H^+-irradiated samples, respectively. Figure 3 shows the current-density dependence of U in (a) pristine and (b) H^+-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$, respectively, constructed by the extended Maley's method. The solid lines indicate power-law fitting to the large J region where the slope in Fig. 2 is positive. Note that deviation of the data from the fitting in the small J region is reasonable since creep is plastic there. The obtained glassy exponents are $\mu = 1.01$ and 1.24 for pristine and H^+-irradiated samples, respectively. For the pristine sample, this value is nearly the same as that obtained in Fig. 2(a), $\mu = 1.09$. On the other hand, the change in μ by H^+ irradiation has the opposite trend. Namely, the value of μ decreases in Fig. 2(b), whereas,

TABLE I. Parameters obtained from the extended Maley's method and the inverse of the plateau value in $S(T)$.

Sample	J_{c0} (MA/cm^2)	U_{00} (K)	μ	$\mu\ \ln(t/t_{\rm eff})$
Pristine	0.85	41.2	1.01	35
H$^+$ irradiated	2.90	93.1	1.24	43

it grows in Fig. 3(b) after H$^+$ irradiation. This is because the vortex system in the H$^+$-irradiated sample crosses over from elastic to plastic creep more gradually as we can see in the main panel and inset of Fig. 2(b). Hence, we may underestimate μ and may overestimate p with the scheme of Fig. 2, and it is more reliable to estimate it from the extended Maley's method of Fig. 3, which uses more data points. For this reason, we conclude that μ is slightly increased by H$^+$ irradiation. Additionally, a slight increase in μ is consistent with the regime of measurement where it is closer to the small bundle regime with $\mu = 3/2$ as we discussed in Fig. 1. Other resultant parameters are summarized in Table I. With these U_{00}, temperature dependence of S is fitted by Eq. (1) with a single free parameter of plateau value $S^{\rm sat} = 1/\mu \ln(t/t_{\rm eff})$ as shown in the inset of Fig. 2. The inverse of this value is also shown in Table I.

Using the parameters obtained above, we calculate J after creep from the attempted function of (true) critical current-density Eq. (6), which is shown as the lower solid line in Fig. 4. In both cases, J is reasonably reproduced, especially at the high J region (i.e., at low temperatures). This means that the present collective-pinning (creep) analysis is appropriate. To get more insight into the pinning mechanisms in IBSs, we also show a function of δT_c and δl pinnings in Fig. 4. These functions are written as $J_c(t)/J_c(0) = (1-t^2)^{7/6}(1+t^2)^{5/6}$ and $(1-t^2)^{5/2}(1+t^2)^{-1/2}$, respectively.[17] From this figure, our model function of J_c can be considered as a superposition of the two pinning mechanisms. To discuss such a mechanism, the generalized inversion scheme (GIS) is utilized.[27,28] Although in this scheme, we have to assume empirical temperature dependence of penetration depth λ and coherence length ξ as $\propto(1-t^4)^{-1/2}$ and $\propto(1+t^2)^{1/2}(1-t^2)^{-1/2}$, respectively, and we can directly reconstruct the true critical current density J_c from J_s and can discuss the pinning mechanism. When we assume $\ln(t/t_{\rm eff}) \sim 23$ in the measurement with field sweeping and choose parameters for the three-dimensional single vortex pinning, J_c is reconstructed as shown in Fig. 4, which is in reasonable agreement with the model function. Similar analyses of a pinning mechanism using GIS in pristine Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ have been attempted in Ref. 29. They also conclude that both δT_c and δl mechanisms are working in this system. Here, we want to compare our work with the similar work by Haberkorn *et al.*[5] In their paper, temperature dependence of (measured) current density $J(T)$ is used to discuss the pinning mechanism in pristine and proton-irradiated Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. However, identification of the pinning mechanism using $J(T)$ is only empirical and lacks a firm physical background. So, although their conclusion and our conclusion on the pinning mechanism are similar, we believe that our identification of the pinning mechanism is more appropriate.

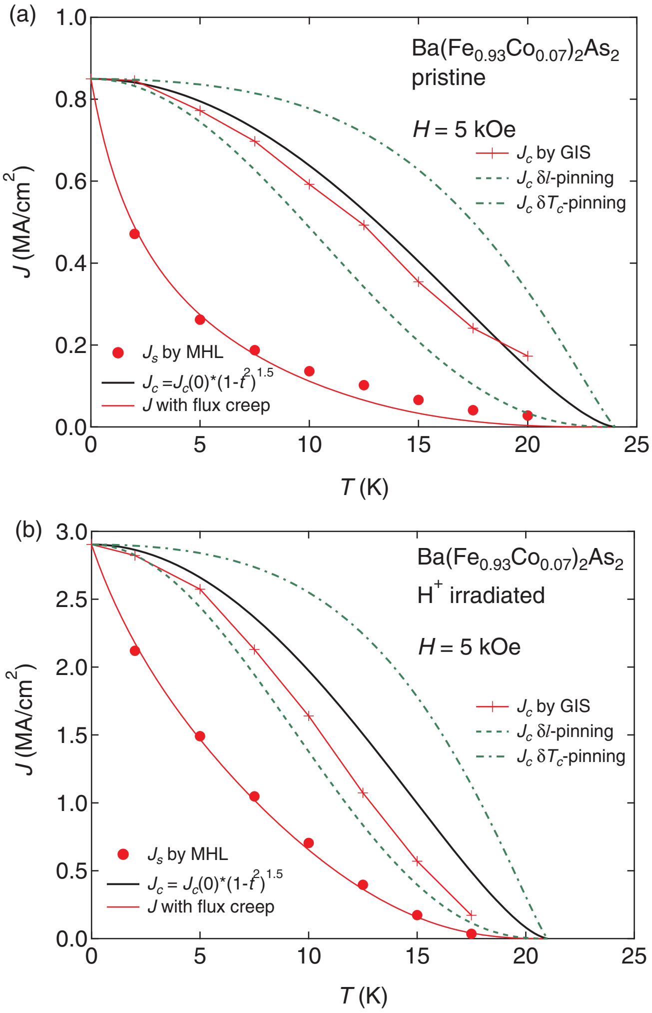

FIG. 4. (Color online) Temperature dependence of J_s determined from the magnetic hysteresis loop (●), model function of J_c before and after creep with the parameters in Table I (solid line), δT_c and δl pinning (dashed line), and J_c reconstructed by GIS (+) in (a) pristine and (b) H$^+$-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$.

We can basically describe the physical quantities for the vortex system by means of collective-creep theory in the cases of both pristine and H$^+$-irradiated Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$. It is noteworthy that the effect of H$^+$ irradiation can be summarized as U_{00} enhancement without replacing model functions $U_0(T)/U_{00}$ and $J_0(T)/J_{00}$. Namely, we can conclude that the effect of H$^+$ irradiation is an enhancement of the collective-pinning force by increasing weak-point-pinning centers without a drastic change in the pinning mechanism.

Finally, we comment on the absolute value of J instead of $J(T)/J(0)$. J is determined by the sum of weak-collective-pinning contribution $J^{\rm wcp}$ and strong-point-pinning contribution $J^{\rm spp}$.[15] Instead, if we assign J to $J^{\rm spp}$ in the absence of flux creep (so that we write J as J_c here), we can estimate the upper limit of the strong-pinning center fraction in the crystal. In the strong-pinning theory,[16] critical current density is written as $J_c \approx 0.14\sqrt{n}\gamma[DF(T)]^{3/2}J_0$. Here, $J_0 = c\phi_0/12\sqrt{3}\pi^2\xi_{ab}\lambda_{ab}^2$ is the depairing current density, $\gamma = H_{c2}^{ab}/H_{c2}^{c}$ is the anisotropy parameter, and n and D are the density and diameter of the pinning centers, respectively. Assuming D as several times of ξ_{ab}, we can simplify $F(T) \approx \ln[1 + D^2/8\xi^2(T)] \approx 1$. Using the pinning center volume $\nu \approx D^3/2$, $n\nu = (J_c/J_0)^2(\sqrt{2} \times 0.14\gamma)^{-2} \approx 0.05\%$ with $\xi_{ab} \sim 34$ Å from $H_{c2}(0) \sim 280$ kOe (Ref. 9) and $\lambda_{ab} \sim 2000$ Å.[30] This value is similar to the value reported in Na-doped $\mathrm{CaFe_2As_2}$.[20]

To summarize, we have studied the effect of proton irradiation up to 1.2×10^{16} $\mathrm{cm^{-2}}$ in optimally Co-doped $\mathrm{BaFe_2As_2}$ single crystals. The critical current density under the self-field is enhanced by a factor of 2.5 at 2 K. The temperature dependence of the critical current density and normalized flux relaxation rate is interpreted by collective-creep theory. With Maley's method, a glassy exponent $\mu \sim 1$ and variation in barrier height for flux creep from $U_{00} \sim 41$ K to 93 K are directly determined. To explain the value of J after the creep from the model function of J_c, J_c is concluded to be controlled by both δT_c and δl pinnings. This model function is consistent with the result of the generalized inversion scheme. The proton-irradiation effect is concluded as doubling of the barrier height in the absence of flux creep.

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