# Mixed state properties of $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ single crystals before and after neutron irradiation

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(Received 2 November 2009; revised manuscript received 16 December 2009; published 20 January 2010)

The reversible and irreversible mixed state parameters of  $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$  (Bi-2223) single crystals were determined by magnetic measurements. One of the samples was exposed to fast neutron radiation in order to investigate the influence of defects introduced into the Bi-2223 matrix. We find that irradiation increases (to a varying extent) the critical current density  $J_c$  over almost the whole field and temperature range analyzed, in particular also at low *B*. This differs from tapes where a  $J_c$  enhancement was only found at elevated fields. Irradiation depth  $\lambda_{ab}$  and the upper critical field  $H_{c2}$ . These two parameters have been calculated with the help of a model that takes thermal fluctuations of vortices into account.  $H_{c2}$ , however, would be expected to decrease according to theoretical predictions for *d*-wave superconductors. The discrepancy is discussed and requires further investigations.

DOI: 10.1103/PhysRevB.81.014516

PACS number(s): 74.25.Ha, 74.25.Sv, 74.62.Dh, 74.25.Op

# I. INTRODUCTION

In the  $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4+\delta}$  (BSCCO) family, where n=1, 2, or 3, the three-layer compound with n=3 (termed Bi-2223 in this paper) is the most interesting superconductor. It shows the highest transition temperature (about 110 K) and critical current density. Bi-2223, however, is only stable in a narrow O<sub>2</sub> pressure and temperature range, which-together with the incongruent melting of its constituents-makes it very hard to synthesize. Only in the last few years different groups succeeded in growing large and phase-pure single crystals with a sharp superconducting transition.<sup>1-3</sup> These high-quality samples finally allow the investigation of intragranular current transport in Bi-2223 and its pinning properties, which is essential to improve the performance of Bibased wires and tapes. In the latter the critical current density  $J_c$  is limited in a large temperature and field range by grain boundaries, rather than by pinning within the grains. It is thus instructive to compare tapes to single crystals, where the effect of grain boundaries is eliminated.

It is particularly interesting to examine the changes in the superconducting properties due to the introduction of crystal defects which act as artificial pinning centers. Neutron irradiation enabled us to do so while having control over the defect density through the neutron fluence.

In this contribution we report on magnetic measurements on Bi-2223 single crystals before and after neutron irradiation. We will give a brief introduction to our experimental setup and the evaluation methods in Sec. II, followed by results on the critical current density, the irreversibility line, and the reversible parameters in Sec. III.

# **II. EXPERIMENTAL**

The magnetic properties of two Bi-2223 single crystals (labeled H5a and W002) grown by the traveling solvent floating zone technique<sup>3</sup> were investigated before and after neutron irradiation. The dimensions of the two samples were between 850 and 1800  $\mu$ m in the crystallographic *ab* plane

and 14 and 21  $\mu$ m in the *c* direction. Their high quality had been confirmed previously by x-ray diffraction measurements performed on samples grown in the same way.<sup>3</sup>

# A. Critical temperature

The critical temperature  $T_c$  was determined from measurements of the magnetic moment vs temperature m(T) in an ac field  $H \parallel c$  (amplitude of 30  $\mu$ T and frequency of 31 Hz) by a Quantum Design 1 T superconducting quantum interference device (SQUID), as shown in Fig. 1.  $T_c$  was obtained as the intersection of two tangents to the slope of the transition and the normal conducting range.

#### B. Hysteresis loops and critical current densities

Magnetic hysteresis loops were measured in an Oxford Instruments vibrating sample magnetometer (VSM) for fields up to 5 T. By approximating the samples as rectangular platelets and using the Bean model,<sup>4</sup> the critical current density for applied fields  $H \parallel c$  was calculated via  $J_c(H) = \{m_i(H)/V\}\{4/[b(1-b/3a)]\}$ .  $m_i$  denotes the irreversible magnetic moment, obtained from moments measured in increasing  $(m_+)$  and decreasing  $(m_-)$  fields via



FIG. 1. (Color online) In-phase and out-of-phase components of the magnetic moment vs temperature in an ac field for sample W002 before irradiation.



FIG. 2. (Color online) Magnetic moment vs temperature at different applied fields  $H \parallel c$  showing the typical crossing point due to thermal fluctuations of vortices for sample W002 before irradiation.

 $m_i = (m_+ - m_-)/2$ , and V is the volume of the sample with dimensions a, b, and c. The induction B was numerically calculated from  $J_c(H)$ , which led to values of  $J_c(B)$ .

### C. Irreversibility line

The irreversibility line was determined from magnetic moment vs temperature m(T) curves for  $H \parallel c$  measured in a Quantum Design 7 T SQUID (see inset of Fig. 6). First the sample was cooled in zero applied magnetic field [zero field cooled (zfc)] and subsequently a field was set and the temperature increased beyond the expected irreversibility temperature  $T_{irr}$  while the magnetic moment was measured in 1 K steps. Immediately afterward a field-cooled (fc) curve down to temperatures below  $T_{irr}$  was obtained. This process was repeated for different fields between 0.1 and 7 T.

Each of these curves showed two distinct features: the drifting apart of the zfc and fc branches at T=T' and a kink at a temperature a few kelvins above, a behavior reported previously for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO) single crystals by Schilling et al.<sup>5</sup> In their study they compared m(T) curves obtained in a SQUID by a conventional method where the sample was moved through the detection coils to the SQUID response voltage when the sample remained at a fixed position. The temperature where the kink was found in m(T) curves acquired by moving the sample corresponded to the irreversibility temperature  $T_{irr}$  obtained by the latter method. They claimed that the data for temperatures below  $T_{irr}$  are affected by field inhomogeneities and therefore do not correspond exactly to the true moment, which leads to the occurrence of the kink. As a consequence in the present work the temperature of the kink (and not the drifting apart of the zfc and fc curves) was taken as the transition from reversible to irreversible behavior at the respective magnetic field.

#### **D.** Reversible properties

The reversible properties were determined using a model developed by Bulaevskii *et al.*<sup>6</sup> which takes into account the entropy contribution to the free energy due to thermal fluctuations of vortices. m(T) curves were, therefore, measured at different  $H \parallel c$  in the 1 T SQUID, which showed the characteristic crossing point at a certain temperature  $T^*$ , as can be seen in Fig. 2. These data were used to calculate  $M(\ln \mu_0 H)$ 

TABLE I. Transition temperatures and widths for the two samples analyzed as well as zero-field critical current densities (before and after neutron irradiation in the case of W002).

Sample	<i>Т</i> <sub>с</sub> (К)	$\Delta T_c$ (K)	$J_c(0 \text{ T}, 10 \text{ K})$ (A m <sup>-2</sup> )
H5a	108.3	3.3	$7.3 \times 10^{9}$
W002 unirradiated	107.6	2.4	$3.00 \times 10^{10}$
W002 irradiated	106.7	2.5	$4.86 \times 10^{10}$

for different temperatures, which was fitted linearly in order to obtain the slope of the magnetization. The penetration depth  $\lambda_{ab}(T)$  was then deduced from  $\partial M/\partial \ln \mu_0 H$  using Eq. (2) from Ref. 7, which is based on the Bulaevskii model. With M(H) and  $\lambda_{ab}(T)$  the upper critical field  $H_{c2}(T)$  parallel to the *c* axis could be obtained via Eq. (6) of the same publication. All other reversible parameters were then deduced from the anisotropic Ginzburg-Landau relations.

#### E. Neutron irradiation

Neutron irradiation of one of our samples (W002) was performed in the central irradiation facility of the TRIGA-MARK-II research reactor in Vienna,<sup>8</sup> during which it was exposed to a fast neutron fluence (E > 0.1 MeV) of  $2 \times 10^{21}$  m<sup>-2</sup>. Studies of YBCO and Bi-2212 single crystals<sup>9,10</sup> showed that this fluence produces defect cascades with a density of  $\sim 1 \times 10^{22}$  m<sup>-3</sup>. They consist of spherical amorphized regions, 2–5 nm in diameter, which corresponds well to the superconducting coherence length in the *ab* plane of Bi-2223 over a broad temperature range. These defects can, thus, act as effective pinning centers. In addition to the cascades smaller defects, like point defects, are induced.

# **III. RESULTS AND DISCUSSION**

### A. Critical temperature

 $T_c$  and the transition widths  $\Delta T_c$  (defined as the temperature difference between 10% and 90% of the Meissner value) before and after irradiation are listed in Table I. Neutron irradiation reduced  $T_c$  of sample W002 by less than 1 K. This decrease, corresponding to 4.5 K per  $10^{22} \text{ m}^{-2}$ , is very similar to the slope found in Ref. 11 for Tl<sub>2</sub>Ca<sub>2</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub> single crystals at *higher* neutron fluences. We did not observe the plateau reported in this study at the lowest fluence analyzed (2×10<sup>21</sup> m<sup>-2</sup>, i.e., the same as in our case). For polycrystal-line (Bi,Pb)-2223 exposed to a neutron fluence of 2.4×10<sup>21</sup> m<sup>-2</sup> a stronger decrease in  $T_c$  by 2 K was found.<sup>12</sup> YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> on the other hand showed a smaller effect of irradiation on the transition temperature with a reduction of only  $1.74 \times 10^{-22}$  K m<sup>2</sup>.<sup>13</sup>

The decrease in  $T_c$  after neutron irradiation has been explained by an increased number of point defects<sup>13</sup> (oxygen vacancies) and is expected from impurity scattering in *d*-wave superconductors.<sup>14</sup> The fact that our sample H5a showed a slightly higher  $T_c$  is, therefore, consistent with it having a lower density of growth-related defects, as indicated by its comparably low value of  $J_c$ .



FIG. 3. (Color online) Magnetic moment vs field (H||c) of sample W002 after irradiation (T=5-50 K, in 5 K steps), together with one curve for the unirradiated sample (10 K). The inset shows the second peak in m(H) at T=35 K, which disappears after irradiation (the irradiated data have been scaled by a factor of 0.1).

# B. Hysteresis loops and critical current densities

Figure 3 shows magnetic hysteresis loops for sample W002 after neutron irradiation measured at different temperatures in the VSM. For comparison a curve measured on the unirradiated sample at 10 K is also depicted to show the increase in magnetic moment due to the introduction of pinning centers by irradiation. From 35 K upward the curves show reversible parts.

The magnetic moment of W002 vs field at T=35 K measured in the 1 T SQUID is presented in the inset of Fig. 3. A second peak is clearly visible at  $\mu_0 H \approx \pm 60$  mT. This phenomenon, which has been explained by an order-disorder phase transition from the low-field Bragg glass to the highfield vortex glass, has been reported previously for Bi-2223 single crystals.<sup>15</sup> The second maximum disappears after irradiation, a behavior which has also been observed in YBCO.<sup>16</sup>

The critical current density vs applied field, which was calculated from VSM m(H) curves as described above, can be seen in Fig. 4. At higher temperatures neutron irradiation increases  $J_c$  over the whole field range. For 10 K, however, the curves for the unirradiated and the irradiated samples merge at a field of about 5 T, and also at 20 K we find a similar trend.

This can be understood as follows. At low fields defect cascades introduced by neutron irradiation act as strong pin-



FIG. 4. (Color online) The critical current densities at different temperatures, which were derived from magnetic hysteresis loops of sample W002 before and after irradiation.

ning centers at all temperatures. As *B* increases we expect them to become less effective; the fluence to which W002 was exposed results in a defect distance similar to the vortex spacing at a field of only 1 T. Nevertheless, we find a strong  $J_c$  enhancement at elevated fields and temperatures. Also, the irreversibility line is shifted to higher values (at least at T>20 K and the fields accessible to our measurements; see below). We conclude that, because of thermal activation, vortices cannot be pinned effectively by the small defects found in the pristine sample in this *B* and *T* range. Defect cascades introduced by neutron irradiation on the other hand act as strong pinning centers, despite their comparably small density.

At low temperatures defects already present in the pristine crystal pin flux lines effectively and the possible increase in  $J_c$  at high *B* due to irradiation is limited. It should be noted, however, that this is only true for the *relative* enhancement. Even at higher fields, like 4 T,  $J_c$  rises by a larger amount at 10 and 20 K, where of course the absolute values are higher, than it does at elevated temperatures. At 5 T we find the peculiar behavior that relative and absolute increases are higher at 20 than at 10 K. This is clearly caused by the growth-related defects being superior to defect cascades at low *T* and high *B*, due to their higher number, hence allowing little room for enhancement at 10 K and 5 T. An increase in  $J_c$  has also been reported after proton irradiation of Bi-2223 single crystals,<sup>17</sup> as well as after neutron irradiation of Bi-2223 and (Bi,Pb)-2223 tapes.<sup>18,19</sup>

The critical current densities of sample W002 before irradiation are slightly higher than those reported for an optimally doped crystal in Ref. 15. The deviations could be explained by variations in oxygen doping and defect densities as well as by the different evaluation formulas used. Our  $J_c$ values match more closely those reported in Ref. 20. The data in both studies<sup>15,20</sup> have been obtained from magnetic measurements on single crystals produced in the same way as our samples.

H5a on the other hand gave smaller values, in particular at low temperatures, which could be explained by a smaller defect density in this pristine sample. Its critical current density vs field at 20 K corresponds well to values obtained from a Bi-2223 tape using SQUID magnetometry (sample 4 in Ref. 21, heat treated at ambient pressure). Even at low fields the tape's  $J_c$  is not suppressed with respect to the single crystal. This implies that over the whole field range the major contribution to its magnetic moment comes from intragranular rather than *inter*granular currents. Consequently also its  $J_c$ , deduced from magnetization measurements, is not limited by grain boundaries at low fields. It needs to be pointed out, however, that the transport  $J_c$  most likely is governed by the properties of the boundaries, not the grains.

At zero applied field and 40 K  $J_c$  of both our unirradiated samples is very similar to the data obtained by transport measurements<sup>22</sup> on a Bi-2223 tape doped with <sup>235</sup>U. Irradiation with thermal neutrons causes the fission of uranium creating randomly oriented linear defects, which seem to have a very similar effect on pinning as defect cascades due to fast neutron irradiation.<sup>18,19</sup> This was also observed in a study conducted on YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> bulk samples<sup>23</sup> and explained by the similar interaction volume of a single flux line with either a collision cascade or a fission track. It is reasonable to expect the same behavior in Bi-2223 since pinning by both kinds of defects should gain the whole condensation energy of a pancake. At low fields the macroscopic critical current density of tapes is, in general, limited by the properties of grain boundaries, rather than grains.<sup>22</sup>  $J_c$ , therefore, is suppressed at low B in tapes after irradiation because of damage incurred by the grain boundaries from neutrons.<sup>18,19,22</sup> The single crystal of course did not suffer from this, which is why it showed a higher  $J_c$  than the tape in this field range after irradiation. At fields above  $\sim 0.5$  T the fission tracks led to an enhancement, however by a significantly smaller amount than was the case after irradiation of our single crystal. Both the pristine and the irradiated tape showed a weaker field dependence; hence, their  $J_c$  exceeds that of the unirradiated single crystal for B > 0.1 T. This behavior can be attributed to the different measurement techniques used and the higher number of defects present in an unirradiated tape compared to a single crystal, which is due to the different production processes. Consequently, the tape surpasses our irradiated sample at fields above  $\sim 1.5$  T.

Chu and McHenry<sup>17</sup> conducted a study of proton irradiation of Bi-2223 single crystals, which leads to the fission of Bi atoms causing columnar damage with a preferential direction parallel to the incident beam.<sup>24</sup> Even aligned defects, however, were found to lead to isotropic pinning;<sup>25</sup> thus, the effect on  $J_c$  of defects caused by fission of Bi can be expected to be akin to those created by fission fragments of  $^{235}$ U. The latter defects in turn have similar pinning properties as defect cascades due to fast neutron irradiation, as mentioned above.<sup>23</sup> In Ref. 17 an increase by a factor of about 3 of self-field  $J_c$  at 20 and 40 K due to proton irradiation is reported. Whereas at the lower temperature we found a similar enhancement factor for our sample, we obtained a value of 13 at 40 K. We attribute this difference to two factors. First Chu and McHenry performed their measurements on an ensemble of 472 tiny single crystals, with their small size leading to a lower self-field and thus a higher  $J_c$ . Consequently, their values are comparable to ours only to a limited extent. Second their samples were grown in molten salt flux of KCl and are supposed to have a large amount of defects and impurities, as confirmed by the wide transition they exhibit. The enhancement factor due to irradiation depends strongly on the number and type of defects in the pristine samples. Bigger defects present before irradiation can lead to an increased  $J_c$  even at elevated temperatures; thus, a smaller enhancement is found.

The dependence of  $J_c$  on temperature is presented in Fig. 5, which illustrates that the relative increase due to irradiation is stronger at higher temperatures. As explained earlier the reason for this is pinning centers in the pristine sample being less effective at higher temperatures, thus allowing more potential for enhancement by irradiation in this regime.

### C. Irreversibility line

Figure 6 shows the irreversibility lines for sample H5a (unirradiated) and for W002 (after irradiation). A few points measured on W002 before irradiation corresponded well to



FIG. 5. (Color online) The temperature dependence of the critical current density of sample W002 for fields of 0.5, 2, and 4 T before and after irradiation.

the values for H5a, thus allowing us to determine the effects of neutron irradiation by comparing H5a to W002 after irradiation.

Not surprisingly the curve for our unirradiated sample replicates well the data of Piriou et al.<sup>15</sup> which were obtained from a very similar optimally doped single crystal by the same method. After neutron irradiation the irreversibility line of W002 shifted to higher values, a behavior reported previously for Bi-2223 tapes<sup>26</sup> and polycrystalline (Bi,Pb)-2223.<sup>12</sup> In the latter study a sample was irradiated to a similar neutron fluence as in the present work and the values for the irreversibility field show qualitatively the same behavior as our data, whereas the absolute values are somewhat higher. This is at least partly due to different evaluation methods employed. The data from Ref. 26 were obtained from transport measurements, which means that they cannot be quantitatively compared to ours. In another study<sup>21</sup> the irreversibility line of a pristine tape was measured using SQUID magnetometry, which led to higher values compared to those of our sample before irradiation. The irreversibility line of their sample 4 is virtually identical over the whole temperature range investigated to the curve for our single crystal after irradiation.<sup>21</sup> Again, at least to a certain extent, we at-



FIG. 6. (Color online) The irreversibility line for samples H5a (unirradiated) and W002 (irradiated) for  $H \parallel c$ . The inset shows m(T) curves of H5a used to determine the irreversibility temperature for respective fields using the kink at  $T_{irr}$ , not the drifting apart of fc and zfc curves at T' (see text).



FIG. 7. (Color online) The penetration depth as measured before and after neutron irradiation. The lines are a fit to the BCS relation with both  $\lambda_{ab}(0)$  and  $T_{c0}$  varied.

tribute these deviations to different evaluation methods, but also to a higher defect density in the pristine tape.

# **D.** Reversible properties

For sample W002 and  $H \parallel c$  the crossing point in the m(T) curves which were used to obtain the reversible properties was found at  $T^*=108.1$  K before and at 107.5 K after irradiation. The penetration depth  $\lambda_{ab}$  was calculated as described above. Figure 7 shows  $1/\lambda_{ab}^2$  plotted vs temperature together with fits to the BCS relation for *d*-wave superconductors,

$$\lambda_{ab}(T) = \lambda_{ab}(0) [1 - (T/T_{c0})^{1.25}]^{-0.47}.$$
 (1)

 $T_{c0}$  is the mean-field transition temperature,<sup>6</sup> compared to the critical temperature  $T_c$  which has been deduced from ac susceptibility measurements (see above). A very good fit was obtained when both  $\lambda_{ab}(0)$  and  $T_{c0}$  were used as fitting parameters. This, however, led to values of  $\lambda_{ab,1}(0)$  and  $T_{c0,1}$ , which are not consistent with Eq. (16) from Ref. 6 and the measured  $T^*$ . We, therefore, used the latter equation to calculate  $T_{c0}$  from  $T^*$  and the interlayer distance s, as determined from the crossing point in m(T), and an estimated value of  $\lambda_{ab}(0)$ . A fit with the so obtained  $T_{c0}$  held constant gave a new  $\lambda_{ab}(0)$ , which was then used to calculate  $T_{c0}$ again. This procedure was repeated until the values remained constant. While this led to very similar values for the penetration depth  $[\lambda_{ab,2}(0)]$ , the transition temperature obtained in this way  $(T_{c0,2})$  appears to be more realistic. The quality of the fit to the measured  $1/\lambda_{ab}^2(T)$ , however, is worse. All values of  $\lambda_{ab}(0)$  and  $T_{c0}$  can be found in Table II.

When the sample was irradiated the penetration depth at T=0 K increased by a factor of about 1.3. This factor can be used to scale the unirradiated to the irradiated data over the whole temperature range analyzed. An increased value of  $\lambda_{ab}$  after neutron irradiation has been reported previously<sup>12</sup> for (Bi,Pb)-2223 and is also consistent with theoretical predictions for the penetration depth in *s*- and *d*-wave superconductors.<sup>27–29</sup> From these models, however, a significantly smaller influence of impurity scattering would be expected (~5% reduction in  $1/\lambda_{ab}^2$ ) than that found in our mea-

TABLE II. Reversible parameters of sample W002 at T=0 K before and after neutron irradiation together with the respective values of  $T_{c0}$  obtained from the fits.  $T^*$  is the temperature of the crossing point in the m(T) curves.

	Unirradiated	Irradiated	
<i>T</i> * (K)	108.1	107.5	
$\lambda_{ab,1}$ (nm)	155	198	
$T_{c0,1}$ (K)	116.9	117.3	
$\lambda_{ab,2}$ (nm)	151	192	
$T_{c0,2}$ (K)	113.3	113.2	
$\mu_0 H_{c2,\text{fit}}$ (T)	113	175	
$T_{c0,H_{c2}}$ (K)	111.8	109.9	
$\mu_0 H_{c2,\mathrm{BCS}}$ (T)	113	190	
$\xi_{ab}$ (nm)	1.7	1.4	
$\mu_0 H_{c,\mathrm{GL}}$ (T)	0.90	0.89	
$\mu_0 H_{c,\text{fit}}$ (T)	0.74	0.70	
$T_{c0,H_c}$ (K)	111.8	111.2	

surements (~40%), given the small decrease in  $T_c$  of our samples after irradiation.

In a next step we deduced the upper critical field, which is depicted in Fig. 8. The data were fitted to

$$H_{c2}(T) = H_{c2}(0) [1 - (T/T_{c0})^{1.5}],$$
(2)

and the fit parameters  $H_{c2,\text{fit}}(0)$  and  $T_{c0,H_{c2}}$  can again be found in Table II. Unlike in the case of  $\lambda_{ab}$  the obtained values for the transition temperature  $T_{c0}$  appear plausible. At 0 K  $H_{c2}$  increased by a factor of about 1.5, which again scales the two curves to each other for all  $T < T_{c0}$ .

Similar values as those obtained from the fit to Eq. (2) were found for the upper critical field at T=0 K using the BCS relation for the clean limit,<sup>30</sup>



FIG. 8. (Color online) Upper critical field vs temperature for sample W002 and  $H \parallel c$ . The data were fitted with both  $H_{c2}(0)$  and  $T_{c0}$  varied.

$$H_{c2,BCS}(0) = -0.73T_c \left. \frac{dH_{c2}}{dT} \right|_{T=T_{c0}},$$
(3)

as can be seen in Table II. The three points closest to  $T_{c0}$  were fitted linearly in order to obtain the slope of  $H_{c2}$  at  $T=T_{c0}$ .

Ossandon *et al.*<sup>12</sup> also reported an increase in the upper critical field after neutron irradiation of the (Bi,Pb)-2223 sample they studied. As in the present work they employed the Bulaevskii model<sup>6</sup> to obtain the reversible parameters and they explained the increases in both  $\lambda_{ab}$  and  $H_{c2}$  by a reduction in the mean-free path for conduction electrons due to radiation damage. We do not believe that our results can be explained by this effect alone. For a 50% increase in  $H_{c2}$ an unrealistically short mean free path  $l \approx 2\xi^{cl}$  would be required according to

$$H_{c2} = H_{c2}^{\rm cl} (1 + \xi^{\rm cl}/l). \tag{4}$$

 $\xi$  is the coherence length and the superscript "cl" indicates the clean limit.

For *d*-wave superconductors a *decrease* in the upper critical field due to impurity scattering was predicted.<sup>31–33</sup> Consequently, these theoretical studies are in contradiction to our findings for  $H_{c2}$ . They were, however, confirmed by experiments, using YBCO samples irradiated by electrons or doped with Zn or Pr.

The discrepancy between our results and the above theories<sup>31–33</sup> can be explained in two ways. The Bulaevskii model only applies to sufficiently anisotropic superconductors, like Bi-2212.<sup>7</sup> The 2223 phase shows a lower anisotropy compared to Bi-2212,<sup>15</sup> and therefore the model might not describe Bi-2223 correctly, potentially leading to unreliable values of  $H_{c2}$ . This would explain why both our data and the results presented in Ref. 12 (which were also deduced from the Bulaevskii model) disagree with theoretical predictions for the upper critical field. It should be pointed out, however, that the  $m(\ln H)$  curves became significantly flatter after irradiation. Linear extrapolation thus leads to higher values of H(m=0), which indicates an increased  $H_{c2}$ , independent of any model.

We therefore conclude that, while impurity scattering as discussed in Refs. 31-33 might also play a role, the strong increase in  $H_{c2}$  must be due to a different mechanism, possibly but not necessarily based on impurity scattering. One explanation would be a reduced Fermi velocity

 $v_F \propto \xi^{cl} \propto 1/\sqrt{H_{c2}}$  after irradiation. This could be due to a decreased charge carrier density or a smearing of the Fermi surface. The first case is unlikely, however, as it would also have a significant impact on  $T_c$ . The latter would make the Fermi surface more isotropic and thus cause a smaller inplane value of  $v_F$ .

We then used the values of  $H_{c2,\text{fit}}(0)$  to calculate the *ab*-plane coherence lengths  $\xi_{ab}(0)$  from the Ginzburg-Landau relation  $\mu_0 H_{c2} = \Phi_0 / 2\pi \xi_{ab}^2$ . After irradiation  $\xi_{ab}(0)$  had decreased by 20%.

The thermodynamic critical field  $H_{c,GL}$  at 0 K followed from the values of  $\xi_{ab}(0)$  and  $\lambda_{ab,2}(0)$  and the Ginzburg-Landau relation  $\mu_0 H_c = \Phi_0 / 2^{3/2} \pi \xi_{ab} \lambda_{ab}$ .  $H_c$  was not changed significantly by irradiation (see Table II). This behavior is expected as  $H_c \propto \Delta(0) \propto T_c$  [where  $\Delta(0)$  is the energy gap at T=0 K] and  $T_c$  has not changed notably. For comparison the thermodynamic critical field was calculated from  $\xi_{ab}$  and  $\lambda_{ab}$ at T>0 K and fitted to

$$H_c(T) = H_c(0) [1 - (T/T_{c0})^2],$$
(5)

which gave  $H_{c,\text{fit}}(0)$  and  $T_{c0,H_c}$ . While  $H_{c,\text{fit}}(0)$  is somewhat smaller than  $H_{c,\text{GL}}(0)$  it again is not significantly affected by radiation damage.

### **IV. CONCLUSIONS AND SUMMARY**

We have conducted magnetic measurements on Bi-2223 single crystals before and after irradiation with fast neutrons. While the transition temperature remained almost constant, we found an increase in the critical current density and the irreversibility line due to the crystal defects introduced by irradiation. This behavior is in agreement with irradiation experiments on BSCCO tapes and single crystals using protons and neutrons.

The reversible properties were determined using a model which takes vortex fluctuations into account. Both the penetration depth and the upper critical field had increased after irradiation. An increase in  $\lambda_{ab}$  is qualitatively consistent with theoretical predictions for impurity scattering in *s*- and *d*-wave superconductors.  $H_{c2}$  in *d*-wave superconductors, on the other hand, would be expected to become lower when the defect density increases. We, therefore, conclude that the introduced defects cause additional phenomena which have a stronger influence on the reversible parameters than that predicted by the above theoretical models alone.

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