Influence of neutron irradiation damage on the equilibrium properties of the polycrystalline $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ superconductor

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Studies of the magnetization of the polycrystalline $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ superconductor revealed substantial changes in several properties, arising from moderate levels of damage from neutron irradiation. In addition to the expected increases in current density (as evidenced by increased magnetic hysteresis), analysis of the equilibrium magnetization revealed significant increases in several more fundamental properties, including the upper critical magnetic field H_{c2} and the London penetration depth λ_{ab}; these increased by 30 and 15 %, respectively, following irradiation with 0.8×10^{17} n/cm^2, with still larger increases at a higher fluence. However, the most intrinsic properties, specifically the thermodynamic critical field H_c and the superconductive condensation energy F_c, were unchanged at the highest neutron fluence used, 2.4×10^{17} n/cm^2. We attribute the changes in fundamental properties to mean-free-path effects, via increased scattering of conduction electrons.

INTRODUCTION

Magnetic studies have been widely used to investigate the properties of high-temperature superconducting (HTSC) materials.[1] For example, in the mixed state, the intragrain persistent current density $J_p(T,H)$ can be found from the *irreversible* magnetization using the critical state (Bean) model. From the *reversible* (equilibrium) magnetization $M(H,T)$, several groups have obtained the London penetration depth $\lambda(T)$ using the London-limit formalism of Kogan, Fang, and Mitra.[2] More recent theoretical developments (Bulaevskii, Ledvij, and Kogan,[3] Kogan *et al.*[4]), have shown that vortex *fluctuations* are important in regions of magnetic field and temperature above the irreversibility line, where vortices are free in principle to move to their equilibrium locations. In particular, the entropy associated with fluctuations introduces a correction to the equilibrium free energy, which modifies the equilibrium magnetization. These modifications are most significant for highly anisotropic, layered materials, and near the superconductive transition temperature T_c. The fluctuation analysis is a useful tool for determining the characteristic magnetic fields and lengths, as well as giving estimates of the superconducting volume fraction of the material. Cho *et al.*[5] showed that this formalism still applies to random polycrystals, if the material is sufficiently anisotropic that only the component of the applied field H perpendicular to the layers is effective. Further below T_c, where fluctuation effects are less significant, the theory of Hao-Clem[6] provides an alternative formulation for determining the fundamental properties of the superconductor from studies of the reversible magnetization in the intermediate field region.[7]

The main objective in this work is to assess the impact of neutron-generated damage on the equilibrium properties of a specific HTSC material. Recent theoretical developments, cited above, provide useful methods for quantifying the characteristic lengths and fields, such as H_{c2} and λ_{ab}. We have used fast neutron irradiation to modify the Bi-based superconductor $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$, which for moderate fluences creates defects in high-T_c superconductors in the form of collision cascades and pointlike defects.[8–10] It appears that the localized damage affects the superconductor in two ways that may be related: (1) it shortens the mean free path for conduction electrons by providing scattering centers, and (2) it increases the (critical and persistent) current density in the material, by increased vortex pinning. Here we study primarily the impact of the mean free path on the equilibrium properties, as there is little information to date regarding these effects in high-T_c superconductors.

EXPERIMENTAL ASPECTS

The subject of this study, the $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ (hereafter BiPb-2223) compound, contains three adjacent CuO layers and is a highly anisotropic superconductor.

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The sample was polycrystalline and sintered. As formed, its diamagnetic onset transition temperature T_c was ≈ 109 K, measured at low magnetic field ($\leq 4-10$ Oe). The aggregate morphology consisted of randomly oriented, thin platelet structures with edge dimensions of ≈ 100 μm. Two pieces cut from the same larger disk were used for comparison; one (mass $=169$ mg) was irradiated with fast neutrons ($E > 0.1$ MeV) at the MIT Research Reactor to increase the density of scattering centers and vortex-pinning sites. The sample was mounted in a polyurethane vial and container. There was no Cd shielding (which is sometimes used to absorb thermal neutrons), due to its excessive heat generation. The sample was cooled by forced air flow, giving an estimated temperature during irradiation of $< 70\,^{\circ}$C. The energy spectrum of fission neutrons has a peak near 1 MeV and extends to ~ 10 MeV.

A neutron fluence $\phi = 0.8 \times 10^{17}$ n/cm^2 led to a threefold increase of irreversible magnetization $M(H,T)$, i.e., increase in the current density for temperatures in the range 5–20 K for all fields. After a total fluence of 2.4×10^{17} n/cm^2, the corresponding enhancements of J_p relative to the unirradiated material were factors of 4–6 in the same temperature range. A more complete discussion of enhancements in current density via irradiation of Bi-Sr-Ca-Cu-O and other materials can be found in a recent review.[11] A second piece (mass$=85$ mg) was left unirradiated for more extensive measurements of the equilibrium magnetization. When unirradiated, the two portions had identical properties, including clear evidence of vortex fluctuation effects in an interval a few K around the low-field T_c.

The magnetic experiments were performed in a multipurpose SQUID- (superconducting quantum interference device) -based magnetometer. Measurements were conducted under identical conditions, using a magnetometer scan length of 2–3 cm, for fixed temperature settings ranging from 40 to 108 K. Before each isothermal measurement, the sample was brought to a temperature well above T_c and then cooled in zero applied field. Once the temperature was stabilized, the magnetic moment "m" was measured at various applied magnetic fields H, increasing from 0.01 to 6.5 T and decreasing back to 0.01 T. To correct for the magnetic moment of the sample holder and any foreign materials, the normal-state magnetic susceptibility was measured for temperatures up to 250 K. It was practically constant, showing no signs of Curie or Curie-Weiss behavior. After correction for the background, the equilibrium (reversible) magnetic moment was determined as the mean value between field-increasing and field-decreasing measurements. In most of the experimental range, however, both signals coincided since the T,H settings were mostly in the reversible region above the irreversibility line of the magnetic phase diagram.

The equilibrium magnetization $M(T,H)$ of the sample was calculated as the reversible magnetic moment per unit of superconducting volume. The determination of the superconducting volume of a sample, however, is nontrivial. Traditionally, the total volume V_{tot} has been obtained by simply dividing the total mass of the sample by the x-ray density (in our case, 6.4 [g/cm^3]). This procedure assumes that all of the BiPb-2223 material is superconducting. The fluctuation analysis indicates, however, that only a fraction of the total volume, the volume ratio "ϵ"$=V_{\text{SC}}/V_{\text{tot}}$, is superconducting. According to the theory outlined below, the volume ratio can be obtained from a specific experimental feature, the field independent magnetization M^* that occurs at the characteristic temperature T^*. The experimental quantities $M^*_{\text{expt}}(=m^*/V_{\text{tot}})$ and T^* are directly observed as a "crossing point" of various $M(T,H)$ lines, as shown in Fig. 1. On the other hand, the theory provides that $M^*_{\text{calc}} = k_B T^*/\phi_0 s$, where "$s$" is the separation between CuO trilayers, k_B = the Boltzmann constant, and ϕ_0 = flux quantum. Since this "crossing point" is an essential feature of the theory, we rescale the total volume by the

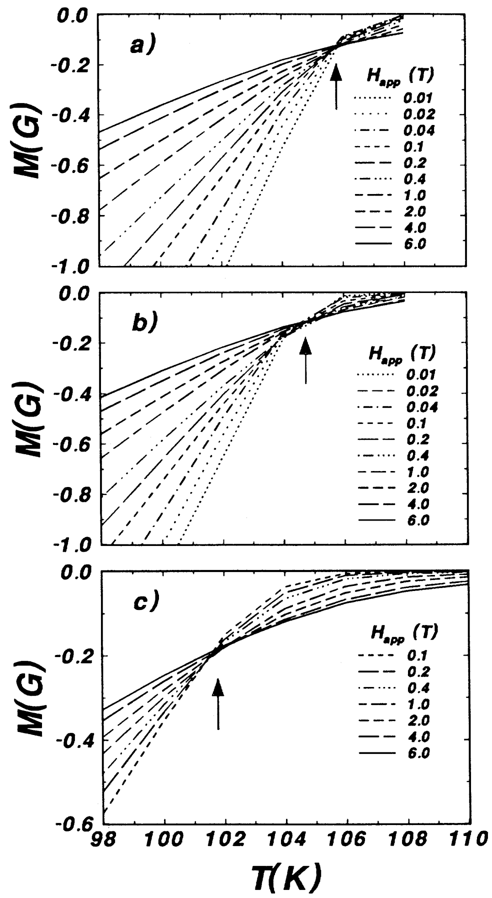

FIG. 1. The mixed state, equilibrium magnetization M of virgin and neutron-irradiated polycrystalline $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ superconductor, plotted vs temperature. Measurements were conducted in various applied magnetic fields H_{app}; the magnetization was independent of field at the crossing point denoted by an arrow. For (a) the neutron fluence was zero (virgin material); for (b) and (c) the fluences were 0.8 and 2.4×10^{17} neutrons/cm^2, respectively. For temperatures above the crossing point, the order of M (top to bottom) is the same as the order in the respective legend (top to bottom); and vice versa below the crossing point.

factor ϵ, with $0<\epsilon<1$, so that the observed and calculated values of M^* coincide. In the present work, ϵ ranged from ~ 0.31 for the virgin sample to ~ 0.45, see Table I.

THEORETICAL FRAMEWORK

Assuming only static interactions between vortices, standard London-limit theory[2] predicts a logarithmic field dependence $M \propto \ln(H)$, where M is the mixed-state reversible magnetization. More recently, Bulaevskii, Ledvij, and Kogan[3] and Kogan *et al.*,[4] have shown that vortex fluctuations contribute to the free energy, due to entropic effects. This modifies the simple London relation by introducing a correction term. With fluctuations included and assuming the magnetic field is applied perpendicular to the CuO layers, one has

$$\frac{\partial M_0}{\partial \ln(H)} = \frac{\phi_0}{32\pi^2\lambda_{ab}^2(T)}[1-g(T)] , \tag{1}$$

where

$$g(T) = 32\pi^2 k_B T \lambda_{ab}^2(T)/(\phi_0^2 s) . \tag{2}$$

The quantity M_0 is the magnetization of a single crystal (or equivalent) with $H \perp$(layers); λ_{ab} is the magnetic penetration depth corresponding to screening by supercurrents in the *ab* planes; and s is the interlayer spacing. For BiPb-2223 containing trilayers of three adjacent CuO sheets, we take s to be the separation of trilayers sets, 1.8 nm. Note that at a characteristic temperature $T^* < T_c$, one has $g(T^*)=1$; consequently M is independent of field and has the value $M^* = k_B T^*/\phi_0 s$. The field independence of the mixed-state magnetization at temperature T^* is an important feature of vortex fluctuation theory as well as nonperturbative scaling theory.[12–14] The results just outlined are valid for temperatures below and approaching $\sim T^*$, near which the correction term markedly affects the deduced values of $\lambda_{ab}(T)$. The free-energy expression leads to the relation

$$\ln\frac{\eta H_{c2}}{eH} = \left[-32\pi^2\lambda_{ab}^2 \frac{M_0}{\phi_0} + g \ln\frac{g}{e}\right]\frac{1}{1-g} , \tag{3}$$

where $e=2.718\ldots$ and the factor $\eta \approx 1.4$ is given by Hao-Clem theory.[6] Equation (3) provides a way for

TABLE I. Results from fluctuation, Hao-Clem, and 2D scaling analyses. Derived magnetic quantities have a $\sim 10\%$ accuracy while temperature values are accurate to within ~ 1 K.

$Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$	Virgin	Irradiated-1	Irradiated-2
Mass (g)	0.085	0.169	0.169
Measured (low field) T_c (K)	109	108	107
Neutron dose (n/cm^2)	0	0.8×10^{17}	2.4×10^{17}
Fluctuation analysis			
M^* (G)	-0.12	-0.14	-0.17
T^* (K)	106	105	102
Volume ratio $\epsilon = V_{SC}/V$	0.31	0.36	0.45
$T_{c0,\lambda}$ (clean limit) (K)	114	113	117
$T_{c0,\lambda}$ (dirty limit) (K)	112	111	113
$\lambda_{ab}(0)$ (clean limit) (nm)	139	152	202
$\lambda_{ab}(0)$ (dirty limit) (nm)	147	160	210
$(dH_{c2}/dT) \approx T_c$ (T/K)	-2.55	-3.3	-6.2
$T_{c0,H}$ (K)	109	110	104
$H_{c2}(0)$ (T)	200	250	460
$\xi_{ab}(0)$ (Å)	12.8	11.4	8.5
$\langle\kappa_c\rangle$ (50–102 K)	84	114	163
$H_c(0)$ (clean limit) (T)	1.31	1.34	1.35
$H_c(0)$ (dirty limit) (T)	1.24	1.28	1.30
Hao-Clem analysis			
(Rescaled for volume fraction ϵ)			
Temperature range (K)	60–90	60–94	60–90
$\langle\kappa\rangle$ (60–90 K)	116	162	270
$(dH_{c2}/dT) \approx T_c$ (T/K)	-3.5	-5.1	-8.4
$T_{c0,H}$ (linear regression) (K)	116	113	108
$H_c(0)$ (BCS-Clem fit) (T)	1.74	1.73	$\equiv 1.73$
$H_{c2}(0)$ (T)	290	410	630
$T_{c,H}$ from Clem's fit (K)	111	110	108
2D scaling analysis			
T_{c0} (K)	109	107	106
$(dH_{c2}/dT) \approx T_c$ (T/K)	-4.0	-5.0	-6.0
$H_{c2}(0)$ (T)	310	380	450

evaluating the upper critical field $H_{c2}(T)$, since all the other quantities are obtained from the preceding analysis.

To this point, the theoretical expressions pertain to the case that $H\perp$(layers), as with a single crystal. According to Cho et al.,[5] these results still apply to a random polycrystal, provided the material is sufficiently anisotropic that only the component of H perpendicular to the layers is effective.[15] Then the magnetization M of a polycrystal is

$$M_s = M_0\langle\cos\theta\rangle + \langle\cos\theta\ln(\cos\theta)\rangle\frac{\phi_0}{32\pi^2\lambda_{ab}^2}(1-g) \ . \qquad (4)$$

The brackets $\langle\cdots\rangle$ denote an angular average with weighting factor $\sin\theta$. For random crystallites, one has $\langle\cos\theta\rangle=\frac{1}{2}$; $\langle\cos\theta\ln\cos\theta\rangle=-\frac{1}{4}$; $M^*=\frac{1}{2}M_0^*$; and $(\partial M/\partial\ln H)=\frac{1}{2}(\partial M_0/\partial\ln H)$. In this work we have used Eqs. (1)–(4) and the weighted angular averages to obtain λ_{ab}, H_{c2}, and related properties.

To complement the fluctuation analysis, the Hao-Clem[6] (HC) formalism was also used to ascertain the effect of neutron irradiation on the superconductor. For internal consistency, the magnetization data were scaled by the volume ratio ϵ deduced above. The HC treatment provides values for the Ginzburg-Landau (GL) parameter $\kappa(T)\equiv\lambda/\xi$ and the thermodynamic critical field $H_c(T)$. The product of these two gives $H_{c2}(T)=\kappa(T)H_c(T)\sqrt{2}$. Also $\lambda(T)$, $\xi(T)$, and $H_{c1}(T)$ can be obtained from the usual GL expressions. Implementing the formalism, however, is not simple, as has been described in detail elsewhere.[7] Generally $H_c(T)$ is relatively well defined (within 3–5 %), while values for κ are less sharply defined (typically within 5–10 %). Consequently the estimates of the upper critical field $H_{c2}(T)$ have uncertainties of 8–15 %. Once values for $H_c(T)$ were determined, $H_c(0)$ at $T=0$ was obtained using the BCS temperature dependence[16] for this quantity. On the other hand, we construct the $H_{c2}(T)$ line near T_c from $H_c(T)$ and κ, and extrapolate H_{c2} to the horizontal axis to define $T_{c,H}$. Finally, the value of $H_{c2}(0)$ at $T=0$ was obtained from the Werthamer-Helfand-Hohenberg (WHH) relation of Werthamer, Helfand, and Hohenberg,[17] $H_{c2}(0)=0.7T_{c,H}(-dH_{c2}/dT)$. The different extrapolation procedures for H_c and H_{c2} are consistent with the fact that the GL parameter κ is temperature dependent.

The third theoretical model used in this work is the two-dimensional scaling theory.[12–14] Assuming a two-dimensional (2D) model for the superconducting transition, the theory predicts that the temperature and field dependence of physical quantities should scale in the variable $t_G=\Xi[T-T_c(H)]/(TH)^{1/2}$, where Ξ is a field- and temperature-independent function and the exponent $\frac{1}{2}$ refers specifically to the 2D case. If $M/(TH)^{1/2}$ is plotted versus t_G, then all $M(T,H)$ curves should collapse onto a single curve, independent of H. For this analysis near T_c, the quantity $T_c(H)$ can be taken as a straight line with slope dH_{c2}/dT and intercept T_{c0}. Scaling to form a universal curve then provides experimental values of T_{c0} and $H_{c2}(T)$.

RESULTS

Figure 1 shows the high-temperature, equilibrium magnetization $M(T;H)$ for the virgin and neutron-damaged BiPb-2223 materials. The various curves denote data for a wide range of magnetic fields, 0.01–6.5 T. Figures 1(a)–1(c) demonstrate the characteristic field independence of the magnetization at temperatures $T^*\approx 106$–102 K for the respective cases. The corresponding magnetization M^* (based on the total volume V_{tot}) at the crossing point is -0.12 to -0.17 G; see Table I for specific values. Note that the fluctuation magnetization in the mixed state persists to temperatures *above* the low field diamagnetic onset T_c of these materials.

Equation (1) predicts a logarithmic dependence of $M\propto\ln H$ on field H. Semilogarithmic plots of M versus $\ln(H)$ at fixed temperatures showed that the data followed this dependence accurately for nearly all temperatures.[18] At the lowest temperatures, some curvature in the semilogarithmic plots developed,[19] as expected; we have excluded these data from the present analysis, which emphasizes the effect of scattering and mean free path on equilibrium superconducting properties. The overall logarithmic field dependence and particularly the presence of the crossing point with field-independent magnetization at T^* are clear indicators of the presence of vortex fluctuations.

To determine the penetration depth $\lambda_{ab}(T)$, we obtained logarithmic slopes $\partial M/\partial\ln H$ as described. Analysis using Eqs. (1) and (2) with appropriate angular and volumetric factors "ϵ" leads directly to λ_{ab} for each isotherm. The results for the virgin and irradiated materials are summarized in Fig. 2 as plots of $1/\lambda_{ab}^2(T)$ versus T. For each case, the solid and dashed lines show the BCS temperature dependencies in the clean and dirty limits, respectively. Points at $T>T^*$, which lie outside of the range of validity of the theory, are plotted for illustration, but were excluded from the fit. From fitting the BCS forms, we extrapolate to obtain $\lambda_{ab}(0)$ and the *mean-field* transition temperature $T_{c0,\lambda}$ at which $(1/\lambda^2)$

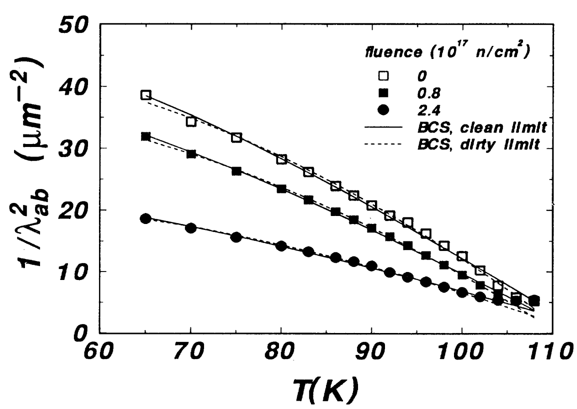

FIG. 2. The temperature dependence of the London penetration depth $\lambda_{ab}(T)$, plotted as $(1/\lambda)^2$. The lines show BCS temperature dependencies in the clean and dirty limits. These results were obtained from fluctuation analyses of the data in Fig. 1.

goes to 0, for the clean and dirty limits; see Table I. At all levels of neutron damage, $T_{c0,\lambda}$ lies several K *above* the low field diamagnetic onset T_c. This is another characteristic feature of the vortex fluctuation theory. The most important result of this analysis, however, is the *systematic increase* in the magnetic penetration depth λ_{ab} with neutron fluence, i.e., damage density. The detailed dependence on fluence will be examined in the discussion section.

The Bulaevskii-Kogan formalism also provides information on H_{c2} at elevated temperatures. So far, we have experimental values for all quantities (λ, g, M_0, etc.) in Eq. (3), except H_{c2}. By plotting the right side of this equation versus $\ln H$ and extrapolating the resulting linear dependence to the horizontal axis, we can obtain $H_{c2}(T)$ to logarithmic accuracy from the intercept. Figure 3(a) shows experimental results for $H_{c2}(T)$ for the virgin and irradiated BiPb-2223 materials. The straight lines in the figure are fitted to the data near but below $\sim T^*$, where the theory is valid. The lines illustrate the linear WHH temperature dependence in the theory of Werthamer, Helfand, and Hohenberg.[17] Note that the slope dH_{c2}/dT ($H \| c$ axis) systematically increases with neutron fluence. Using the WHH relation $H_{c2}(0) = -0.7 \times (dH_{c2}/dT) T_{c0}$ gives values for $H_{c2}(0)$ increasing from ~ 200 to 450 T. From these results, we compute the *ab*-plane coherence lengths $\xi_{ab}(0)$ from the standard Ginzburg-Landau relation $H_{c2} = \phi_0 / 2\pi\xi_{ab}^2 = 2^{1/2}\kappa H_c$. This shows that $\xi_{ab}(0)$ decreases from 12.8 to 11.4 to 8.5 Å, respectively; i.e., an overall reduction of about 34%.

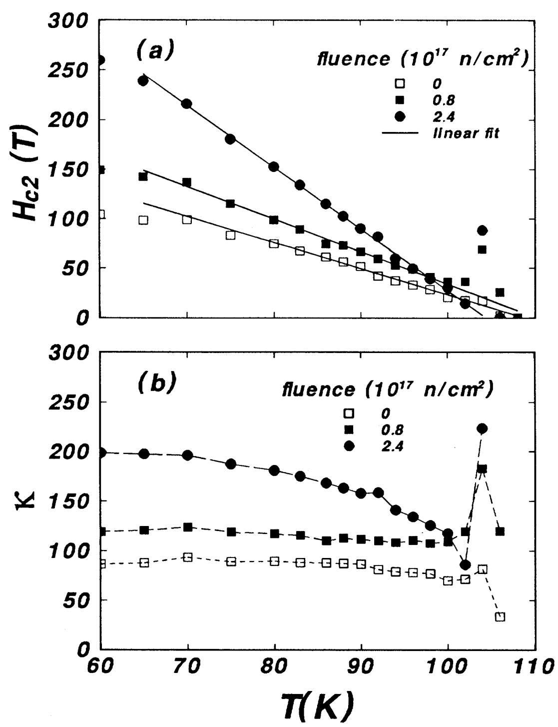

FIG. 3. The dependence on temperature of (a) the upper critical magnetic field H_{c2} ($H \| c$ axes) and (b) the Ginzburg-Landau parameter $\kappa = (\lambda_{ab}/\xi_{ab})$ for virgin and neutron-irradiated BiPb-2223. As for Fig. 2, a fluctuation analysis was used. Note that H_{c2} varies approximately linear with temperature near T_c, while κ is nearly constant, as expected.

In light of these marked changes, it is very instructive to examine the thermodynamic critical field $H_c(0) = \phi_0 / 2^{3/2}\pi\xi_{ab}\lambda_{ab}$, since it is intimately related to the fundamental energetics of the superconductor. As shown in Table I, the values obtained for this quantity are very similar, $H_c(0) \approx 1.3$ T, for the virgin and both irradiated samples. Thus the condensation energy $H_c^2/8\pi$ was not altered by radiation damage, which is consistent with the small change in T_c ($\pm 2\%$) and the fact that it is an intrinsic thermodynamic property.

Figure 3(b) compares the Ginzburg-Landau parameter $\kappa_c = \lambda_{ab}/\xi_{ab}$ for the three samples. As expected, the experimental values of κ are nearly temperature independent. For each sample, Table I lists the average value of κ for the temperature range 50–102 K. The progressive increase of κ with neutron fluence ϕ is consistent with the observed increase in H_{c2} and constancy of the thermodynamic critical field H_c. In contrast with the nominal temperature independence of κ seen in Fig. 3(b), the values obtained from any analysis neglecting fluctuation effects ($g = 0$) are unphysical near T^*, with κ tending to diverge as T increases.

The fluctuation analysis gives evidence that irradiation-generated disorder produces remarkable changes in many superconducting properties. To corroborate these findings, we used the independent Hao-Clem[6] formalism to reanalyze the background-corrected, equilibrium magnetization data for all samples. Since this technique neglects thermal fluctuations of vortices, the results near T_c are not reliable; hence we concentrate on the lower temperature range from 60 to ~ 90 K. Also, we base the magnetization on the superconducting volume of each sample, as obtained from the fluctuation analysis, to facilitate numerical comparison. The resulting values are tabulated in Table I. The results confirm the significant increases of κ and the H_{c2} slope due to irradiation damage. Consistent with our previous finding, the thermodynamic critical field $H_c(0) = 1.7$ T was unchanged by the first irradiation with 0.8×10^{17} n/cm^2. For unknown reasons, the numerical solutions to the Hao-Clem equations converged poorly for the higher fluence sample, with excessive interaction between the trial values of κ and H_c. To proceed with this analysis, we set $H_c \equiv 1.7$ T as obtained for the lower irradiation levels; this plausible assumption is consistent with the results of the fluctuation analysis. The resulting values for this irradiation level are shown in the Hao-Clem section of Table I.

We now consider a third analytical approach, a scaling analysis. According to the theory outlined above, a plot of $M(H,T)/(TH)^n$ versus $[T - T_c(H)]/(TH)^n$ should collapse onto a single curve; for 2D scaling as is appropriate for highly layered materials, the exponent $n = \frac{1}{2}$. The temperature $T_c(H)$ can be taken as a linear function of temperature, with slope dH_{c2}/dT and intercept T_{c0} on the T axis. Adjusting these parameters yielded the scaling illustrated in Fig. 4. The figure includes results for

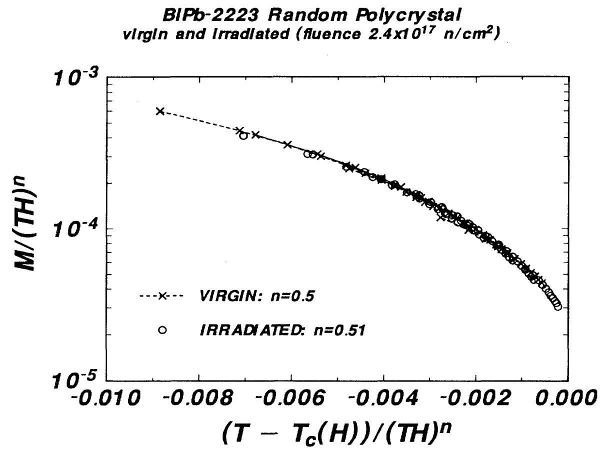

FIG. 4. Scaling analysis of the experiment magnetization for a 2D superconductor. The results properly collapse onto a single "universal" curve, using the critical field slopes and parameter values shown in Table I. The temperature interval treated is 100–106 K for the virgin sample and 96–104 K for the sample irradiated with 2.4×10^{17} n/cm^2. Measurements for the intermediate fluence of 0.8×10^{17} are very similar, but for clarity are not shown.

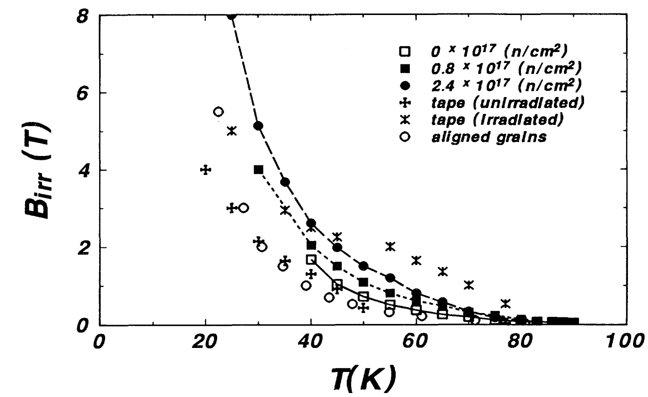

FIG. 5. The irreversibility line $B_{\mathrm{irr}}(T)$ for the virgin and neutron-damaged materials. Generally the irreversibility field increases with damage density. For comparison, the figure includes results for aligned grains of BiPb-2223 (Ref. 21) and for textured tape with and without columnar defects (Ref. 20).

both the virgin sample and the sample irradiated with 2.4×10^{17} n/cm^2. The respective parameter values are $n=0.50$, $T_{c0}=109$ K, and slope $=-4$ T/K for the virgin sample, compared with $n=0.51$ and 106 K, and -6 T/K for the more heavily irradiated sample. For clarity, the results for the intermediate irradiation are not shown in the figure. However, the data overlay those shown, with parameter values $n=0.51$ and 107.8 K, and slope -5 T/K. These results again point to significant increases in H_{c2} upon irradiation.

In addition to the documented changes in the characteristic fields and lengths generated by irradiation, we find that the irreversibility line $B_{\mathrm{irr}}(T)$ increases notably with neutron damage. This line in the B-T phase diagram defines two regions, one at low temperatures with a finite persistent current density J_p and a second at higher temperatures with immeasurably small J_p. Experimental results are shown in Fig. 5. While the nature and origin of this line continue to be highly controversial, particularly in the presence of disorder, we concern ourselves here with establishing the changes in B_{irr} associated with neutron-generated defects. Experimentally, we obtained values for B_{irr} from plots of the pinning force density J_pH versus field H, for fixed temperature T. As the field H increases from zero, the pinning force increases, passes through a peak, then decreases nearly *linearly,* and finally approaches the horizontal axis asymptotically. We extrapolate the characteristic linear decrease to the axis, where $J_pH=0$, and use the intercept to demarcate B_{irr}. Operationally, this procedure is often better defined than the alternative procedure of trying to find the point at which the hysteresis $\Delta M \propto J_p$ in a magnetization curve disappears. Applying the same procedure to the virgin and irradiated materials gives values for $B_{\mathrm{irr}}(T)$ that generally increase with neutron fluence.

For comparison, Fig. 5 includes other measurements of the irreversibility line in this compound.[20,21] The data of Li *et al.*[21] correspond to unirradiated aligned powder of transverse dimension ~ 10 μm. Also shown are measurements of Civale *et al.*[20] on textured BiPb-2223/Ag tape, both as prepared and irradiated with 1-GeV Au ions to form columnar defects. The irreversibility fields for all the unirradiated materials are roughly consistent, although some dependence on sample dimension has been demonstrated.[21] With neutron irradiation, B_{irr} steadily increased, as observed[22] previously in single crystals of $Bi_2Sr_2Ca_1Cu_2O_8$. This is likely due to increased vortex pinning from collision cascades and associated defects. The highest irreversibility line belongs to material with columnar defects, which provide the most effective vortex pinning sites known. Indeed, quite substantial expansions of the irreversible region have been demonstrated for the sister compound $Bi_2Sr_2Ca_1Cu_2O_8$, both with aligned columnar defect[23,24] and with randomly oriented (splayed) columnar defects produced by GeV energy protons.[25] For the BiPb-2223 material, then, the irreversibility field progresses systematically: it is lowest in unirradiated materials with only naturally occurring defects; it increases with the density of neutron-generated damage; and it is highest in the presence of columnar defects.

DISCUSSION

This work has established that several of the characteristic fields and lengths of BiPb-2223 superconductor are substantially modified by neutron-irradiation-induced damage. Using three independent theoretical models, we observe consistent trends with neutron fluence, specifically increases in the upper critical magnetic field $H_{c2}(0)$ and the London penetration depth λ. The fluctuation analysis indicates that the slope of H_{c2} and its calculated value at zero increase by factors of 1.3 and 2.3 relative to the unirradiated material, for respective fluences of 0.8 and 2.4×10^{17} neutrons/cm^2. The Hao-Clem analysis yielded similar factors of 1.46 and 2.4 for the corresponding cases. Also, 2D scaling yielded increases in H_{c2} slope of 1.25 and 1.5 for the respective irradiations. The penetration depth increased significantly for the two fluences, by 10 and 45% relative to the virgin material.

This work shows that localized neutron damage causes substantial changes in the equilibrium properties of this high-T_c superconductor. Three different analyses all point to changes in H_{c2} by similar factors. In comparing results from the three methods, we focus on relative changes. It is perhaps not surprising that the numerical values from the different analyses differ, which can arise from dissimilar angular averages in the polycrystalline material. However, (portions of) the same polycrystal was used throughout the study, so any theory-specific angular average should be the same for all fluences, i.e., each specific analysis is internally consistent. In contrast with H_{c2} and λ, the thermodynamic critical field H_c did *not* change appreciably, but remained remarkably constant. This provides an important check on the study, for it should not change: with moderate levels of neutron damage and a slight depression of the low field T_c from 109 to 107 K, one expects only negligible changes in the average order parameter and in the condensation energy.

What microscopic phenomenon can explain the clearly documented increases in H_{c2} and London penetration λ? A likely mechanism is a reduction in the electronic mean free path due to irradiation damage. Monte Carlo calculations and transmission electron microscopy studies[8,9] of other high-T_c materials with moderate levels of neutron damage show the formation of collision cascades, which will be accompanied by pointlike defects. Certainly such defects produce electronic scattering, a reduced mean free path, and increased intragrain electrical resistivity ρ in the normal state. For 3D superconductors, there is a well-developed theory predicting that the slope of H_{c2} increases as the mean free path decreases.[26] This theory has been supported by experiments[26] on three-dimensional superconductors, such as the $A15$ materials V_3Si and Nb_3Sn. Unfortunately, no corresponding theory for highly anisotropic superconductors exists, and intergranular electrical resistance in our polycrystalline BiPb-2223 precluded meaningful measurement of the intragrain resistivity. Moreover, it is likely that defects in this layered, 2D material affect the electronic system more strongly than for 3D materials, since there is less phase space for conduction electrons to scatter into.

Recognizing the limitations just listed, we now apply the available 3D theory to the current results. A convenient tabulation of theoretical relations has been given by Orlando *et al.*,[26] whose notation we use in the following. For example, one has

$$H_c = 4.23\gamma^{1/2}T_c(1-t) , \tag{5}$$

where $t = T/T_c$ is the reduced temperature, γ is the coefficient of the linear electronic heat capacity, magnetic fields are measured in Oe, and all lengths are measured in cm. With the measured value of H_c, this relation predicts $\gamma \approx 900$ erg/cm^3 K^2. This estimate compares reasonably with the value $\gamma_{\rm BCS} \sim 600$ erg/cm^3 K^2 for BiPb-2223 deduced by Schnelle *et al.*[27] from the jump in specific heat near T_c. Considering the slope of H_{c2} near T_c, we assume that the clean limit applies for the virgin sample, since ξ_{ab} is short. In this limit, the 3D theory provides that

$$-dH_{c2}/dT = 9.55\times 10^{24}\gamma^2 T_c(n^{2/3}S/S_F)^{-2} , \tag{6}$$

where n is the density of conduction electrons, S is the Fermi-surface area, and S_F is the Fermi-surface area for a free-electron gas of density n. Using the measured slope of H_{c2}, -2.6×10^4 G/K, one finds $(n^{2/3}S/S_F) \sim 2 \times 10^{14}/\mathrm{cm}^2$; setting $S/S_F = 1$ gives the crude estimate that $n \sim 2\times 10^{21}$ cm^{-3}, which is not unreasonable. The London penetration depth λ gives a consistency check through the relation

$$\lambda(T=0) = 1.33\times 10^8\gamma^{1/2}(n^{2/3}S/S_F)^{-1} . \tag{7}$$

This gives the value $\lambda \sim 220$ nm, which compares reasonably with the measurement of $\lambda = 139$ nm.

After irradiation, the mean free path l is shorter. Then it is appropriate to use the full expression for the slope of H_{c2}:

$$-dH_{c2}/dT = [9.55\times 10^{24}\gamma^2 T_c(n^{2/3}S/S_F)^{-2} + 6.67\times 10^8\gamma(1/l_{\rm tr})(n^{2/3}S/S_F)^{-1}] , \tag{8}$$

$$-dH_{c2}/dT = [9.55\times 10^{24}\gamma^2 T_c(n^{2/3}S/S_F)^{-2}[1+0.88(\xi_0/l_{\rm tr})] . \tag{9}$$

Here $\xi_0 = 7.95\times 10^{-17}(n^{2/3}S/S_F)/(\gamma T_c)$ is the BCS coherence length (in the ab direction, in this case); and the prefactor in Eq. (9) is the slope of H_{c2} in the clean limit. (For these purposes, we ignore factors of order unity, most notably the Gorkov function $R(\lambda_{\rm tr})$, where $1 \le R(\lambda_{\rm tr}) \le 1.17$.) From Eq. (8), we obtain the estimate that $l_{\rm tr} \sim 9$ A following the second irradiation, i.e., the mean free path becomes the order of the ξ_{ab} and is only a small multiple of the crystallographic unit-cell length. Of course, the numerical values are no more than estimates, but the resulting values are surprisingly reasonable.

A qualitative comparison of these results with the mean spacing between defects is instructive. To estimate the defect density, we first find the number pervolume $(N/V)_{\rm coll}$ of neutron-atom collisions. We have $(N/V)_{\rm coll} = (N/V)_{\rm atoms}\phi\sigma \approx 3\times 10^{16}$ cm^{-3}, where $(N/V)_{\rm atoms} = 7\times 10^{22}$ atoms/cm^3 is the atomic number density of Bi-2223, $\phi = 2.4\times 10^{17}$ n/cm^2 is the higher neutron fluence, and $\sigma \approx 2\times 10^{-24}$ cm^2 is an average nuclear cross section giving recoil energies ≥ 30 keV, which is the energy required to produce collision cascades.[28] The recoiling ions create localized vacancies, interstitials, and extended collision cascades. Using the computer code TRIM,[29] we approximate the ion damage with that produced by 50-keV Kr ions;[28] these ions produce cascades in $YBa_2Cu_3O_7$ crystals that are similar to neutron-generated cascades. The Monte Carlo calculations show that each ion creates approximately 900 vacancies; irradiation with other "equivalent ions," e.g., Cr whose atomic number is the weighted average of the constituents in $Bi_2Sr_2Ca_2Cu_3O_{10}$, gives similar numbers of vacancies. Hence the calculated density of vacancies is $\sim 3\times 10^{19}$ cm^{-3} (which may be an underestimate, since it neglects lower energy recoils). This corresponds to a mean vacancy separation of ~ 30 A, which is comparable in magnitude to the estimate for mean free path l_{tr}. These crude analyses strongly suggest that mean-free-path effects can play a significant role in modifying the characteristic lengths and fields in suitably modified high-T_c superconductors.

The dependence on neutron fluence is interesting. For moderate levels of neutron damage, one can expect an inverse variation in the mean free path, with $(1/l_{tr}) \propto$ (neutron fluence ϕ); recent work[28] has shown that the measurable defect density scales linearly with neutron fluence. Reference to Eqs. (8) and (9) then suggests that the slope of H_{c2} should increase approximately linearly with fluence. Indeed, Fig. 6(a) shows just such a linear dependence for the H_{c2} slopes near T_c, as obtained from the fluctuation and Hao-Clem analyses. The straight lines are linear fits to the experimental data. Figure 6(b) shows the dependence on fluence for the magnetic penetration depth. According to the 3D theory, this quantity depends on the mean free path in the form $\lambda_{ab}^2 \propto [1+0.88(\xi_0/l_{tr})]$. Indeed, Fig. 6(b) shows an approximately linear variation with fluence, as provided theoretically. Note, also, that the expressions for λ_{ab}^2 and for dH_{c2}/dT [Eq. (9)] have the *same* functional form $\propto [1+0.88(\xi_0/l_{tr})]$ with different prefactors. To facilitate comparison of dependencies on fluence ϕ, Fig. 6(c) presents the experimental data (symbols) and linear regressions (lines), scaled in each case by the corresponding fitted value at $\phi=0$. Consequently, at zero fluence, all fit lines pass through unity by construction; the experimental points lie close to unity, with some experimental scatter. The major point is that the normalized data in Fig. 6(c) have very similar slopes, i.e., very similar dependencies on l_{tr}, as predicted theoretically. These consistencies in this set of results further support the idea that localized damage affects the equilibrium superconducting properties via mean-free-path effects.

Recent work by Griessen *et al.*[30] has shown that mean-free-path effects can significantly affect the nonequilibrium (hysteretic) properties of high-T_c superconductors. In particular, they argue that the vortex pinning in a series of YBaCuO thin films is due to *scattering* from naturally occurring pointlike defects, rather than localized depressions of T_c.

In conclusion, this work shows that neutron-generated damage produces increases in several of the equilibrium properties of a specific high-T_c superconductor, $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$. For the moderate levels of neutron damage used, several independent analyses point to significant increases in the magnetic penetration depth λ_{ab} and upper critical magnetic field H_{c2}, but no change in the thermodynamic critical field H_c. The results appear qualitatively consistent with well-established 3D isotropic theory for the effects of mean free path on superconducting properties. Overall, it appears that mean-free-path effects can modify the properties of high-T_c superconductors substantially. Further investigations are underway to explore these phenomena.

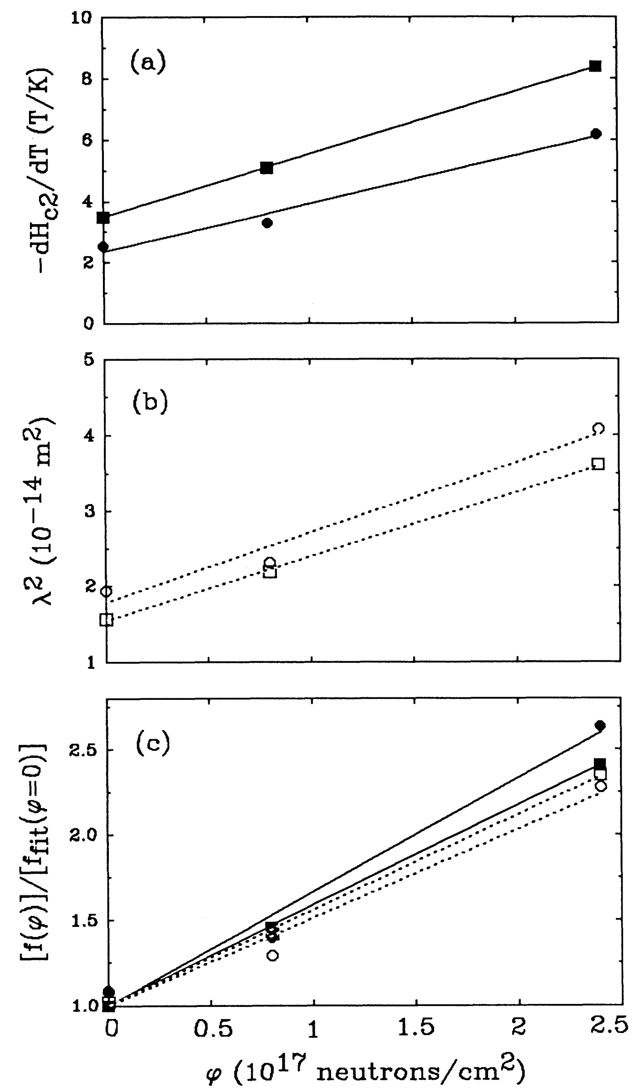

FIG. 6. Dependence of the experimental quantities $f(\phi)$ on neutron fluence ϕ, where $f(\phi)$ is (a) the slope of the upper critical magnetic field $-dH_{c2}/dT$ and (b) the square of the magnetic penetration depth λ_{ab}, extrapolated to $T=0$. Straight lines are linear regressions to illustrate the theoretical form; discrete symbols are results from the Hao-Clem data analysis (■,□) and the fluctuation analysis (●,○). In (c), these results are normalized by the respective fitted values at $\phi=0$, $f_{fit}(\phi=0)$; the slopes are similar, which reasonably approximates the theoretical prediction that they are identical.

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