INSTITUTE OF PHYSICS PUBLISHING SUPERCONDUCTOR SCIENCE AND TECHNOLOGY

Supercond. Sci. Technol. **14** (2001) 666–671 PII: S0953-2048(01)26343-X

Influence of randomly oriented columnar defects on the irreversible and reversible magnetization of $Tl_2Ba_2CaCu_2O_x$ superconductor

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Received 4 June 2001
Published 16 August 2001
Online at stacks.iop.org/SUST/14/666

Abstract
Bulk polycrystalline $Tl_2Ba_2CaCu_2O_x$ materials were irradiated with 0.8 GeV protons to introduce randomly oriented columnar defects. Proton fluences up to 8.7×10^{20} m^{-2} were used to create defect arrays corresponding to a 'matching field' of 1.5 T. Studies were conducted on the superconductive transition temperature, the Meissner fraction, the intragrain persistent current density against the magnetic field and temperature, and the equilibrium magnetization. The magnetization was modelled using London theory with the addition of vortex–defect interactions, yielding physically reasonable parameters.

1. Introduction

To maximize the current-carrying capability of high-T_c superconductors in large magnetic fields, strong pinning of vortices is necessary. It is generally accepted that some form of correlated disorder is most effective. Columnar defects (CDs) which have been widely studied, are typically formed by irradiation with energetic heavy ions that are highly ionizing and create tracks of amorphous material along their path [1]. Defect arrays with parallel tracks are most familiar, although many other configurations (inclined defects, crossed arrays with 'designer splay', etc) have been investigated. One problem with heavy ions is their limited range, typically a few tens of micrometres. To overcome this limitation, Krusin-Elbaum *et al* [2] devised a method to use 0.8 GeV protons to form CDs indirectly, via a fission process. The advantage of the lighter particles is their large range, greater than 0.5 m in cuprate superconductors. In this process, the energetic proton is absorbed by a heavy nucleus that is a natural constituent of the superconductor—thallium in the present case. The impact of the proton causes spallation of several neutrons and protons and energizes the nucleus into a highly excited state. The nucleus fissions into two particles of roughly equal mass, each with ∼100 MeV energy. These recoiling particles, created deep within the superconductor, form randomly oriented CDs. In a different approach, to form pins deep inside a material, Schultz *et al* [3] created CDs by introducing ^{235}U into the superconductor, which is induced to fission by irradiation with thermal neutrons.

The impact of proton-generated fission defects was first demonstrated in magnetic studies of Bi-2212/Ag tapes [2]. Subsequently Safar *et al* [4] showed that the process enhances the transport current density in Bi-2223 prototype materials. The potential for processing larger structures was demonstrated by the successful irradiation of an entire prefabricated coil of BiSrCaCuO tape, resulting in an increased current-carrying capability [5]. More recently, we have shown that the proton-induced fission process is generally applicable in cuprate superconductors that contain a sufficient density of heavy nuclei [6]. A variety of interesting physical phenomena have been observed in materials containing randomly oriented CDs. These include the presence of significant temperature independent quantum tunnelling of vortices in B-2212 materials [7]. Another finding [8] was that sufficiently high superconductive anisotropy can lead to a rescaling and 'refocusing' of the splayed landscape of random CDs, even in polycrystalline materials, which is a feature that will be used in a later discussion.

In the present study, we investigate the impact of gigaelectronvolt proton irradiation on the superconductive properties of $Tl_2Ba_2CaCu_2O_x$ superconductors. Features for investigation include the transition temperature T_c, the Meissner (flux expulsion) fraction, the intragrain persistent current density J as obtained from the magnetization, establishment of the optimum defect density, and study and analysis of the changes in the mixed state equilibrium magnetization resulting from interactions between vortices and randomly oriented columnar defects.

2. Experimental aspects

The study was conducted on bulk polycrystalline Tl_2Ba_2Ca-Cu_2O_x materials. The crystalline unit cell is tetragonal, with lattice constants $a = 0.385\,50$ nm and $c = 2.9318$ nm (cell volume of 2.18×10^{-28} m^3) containing two sets of adjacent oxygen–copper layers. Rectangular slabs of 3×3 mm^2 cross section and $\sim$1 mm thickness were all cut from the same original pellet. The mechanical density of the virgin sample was measured to be 6.2 kg m^{-3}, compared with the density of 7.458 kg m^{-3} calculated from the x-ray lattice parameters and the ideal stoichiometry, this means that the material was 83% dense. The samples were irradiated at room temperature in air with 0.8 GeV protons at the Los Alamos National Laboratory. Proton fluences φ_p, as determined from the activation of Al dosimetry foils, were 0, 0.41, 0.74, 0.92, 1.9, 3.5, and 8.7, all in units of 10^{20} protons m^{-2}. The energetic protons have a significant cross section of $\sigma_f \simeq 110$ mb to induce the prompt fission of the heavy thallium nuclei. The resulting two recoil fragments, each with an average energy of $\sim$100 MeV, form CDs of average length $\sim$8 μm. Since the fission process is random in direction, the CDs are randomly oriented in space. The density of fission events is given by $N/V = \varphi_p \sigma_f n_{Tl}$, where n_{Tl} is the number density of Tl nuclei. Since each fission produces one CD, this also gives the volume density of defects. One can convert this into an approximate area density of CDs by multiplying by the track length, and the area density can be expressed in units of a matching field B_Φ by multiplying by the flux quantum Φ_0. Hence the corresponding defect densities are $B_\Phi = 0$, 0.08, 0.14, 0.18, 0.37, 1.0, and 1.5 T, respectively. Previous transmission electron microscopy (TEM) studies of similar materials (Bi-2212 and Hg-cuprates) [2, 9] have demonstrated the presence of randomly oriented columnar defects in these high-T_c materials, and shown the generality of the fission process in forming randomly oriented CDs in HTSs containing heavy (Hg, Tl, Pb, Bi, etc) nuclei.

The superconductive properties of the virgin and irradiated materials were investigated magnetically. A SQUID-based magnetometer (model Quantum Design MPMS-7), equipped with a high homogeneity 7 T superconductive magnet, was used for studies in the temperature range 5–300 K, in applied fields of up to 6.5 T. To determine the superconductive transition temperature T_c of the samples, the magnet was 'reset' by heating it above its T_c to release trapped flux, then a field of 4 Oe (0.4 mT) was applied at a sample temperature of 5 K. The temperature was swept to 120 K, then the sample was cooled in the same field to determine the (normalized) Meissner fraction $-M/H$; if all flux were expelled from the sample, then this (SI) quantity is +1. The results of this low-field study are shown in figure 1 and discussed below.

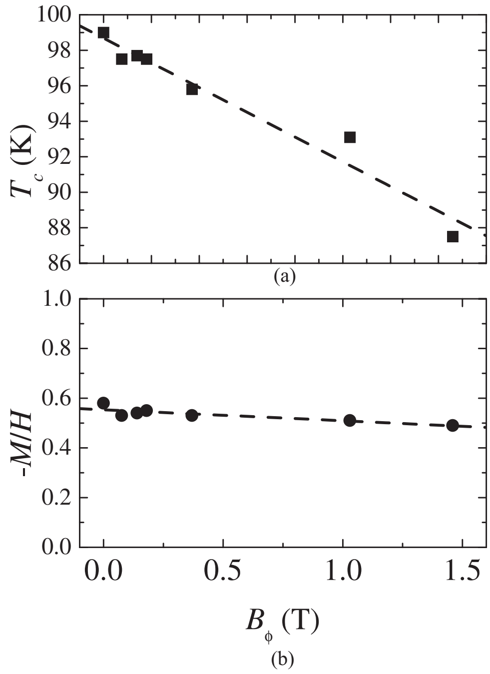

Figure 1. (a) The superconductive transition temperature (onset) and (b) the normalized Meissner signal $-M/H$, plotted against proton fluence expressed as an equivalent 'matching field B_Φ' density of randomly oriented CDs. Measurements were conducted in an applied field of 0.4 mT.

The isothermal magnetization M was measured as a function of the applied magnetic field. Below T_c and below the irreversibility line, the magnetization was hysteretic due to the presence of intragrain persistent currents. From the magnetic irreversibility $\Delta M = [M(H\downarrow) - M(H\uparrow)]$, the persistent current density J was obtained using the Bean critical state relation $J \propto \Delta M/r$, where $r = 4$ μm is the mean grain radius. In the normal state above T_c, the magnetic response was temperature dependent and accurately described by a Curie–Weiss law. We used this dependence to correct for the background paramagnetism below T_c, in order to isolate the equilibrium magnetization M_{eq} in the superconductive state. Above the irreversibility line where $\Delta M = 0$, this process yields M_{eq} directly; near the irreversibility line where ΔM is small, we obtain M_{eq} from the (background-corrected) average magnetization $= [M(H\downarrow) + M(H\uparrow)]/2$.

3. Experimental results

The process of doping the superconductor with CDs locally destroys a portion of the material. This serves to depress somewhat the overall order parameter and this is reflected in the transition temperature T_c, which is shown as a function of proton fluence (expressed as a matching field) in figure 1(a).

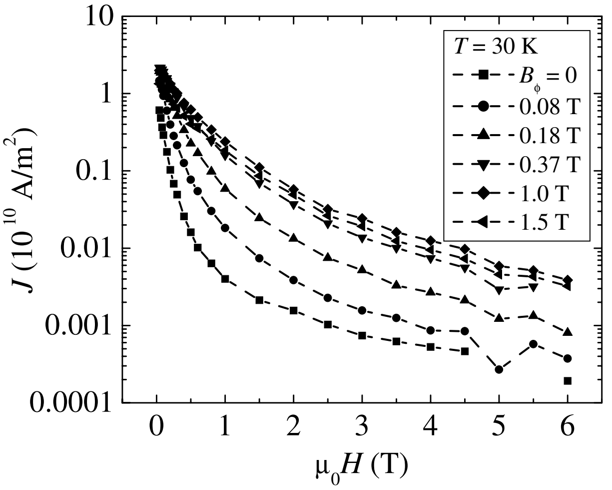

Figure 2. The persistent current density J against applied magnetic field, for various irradiated materials at a temperature $T = 30$ K.

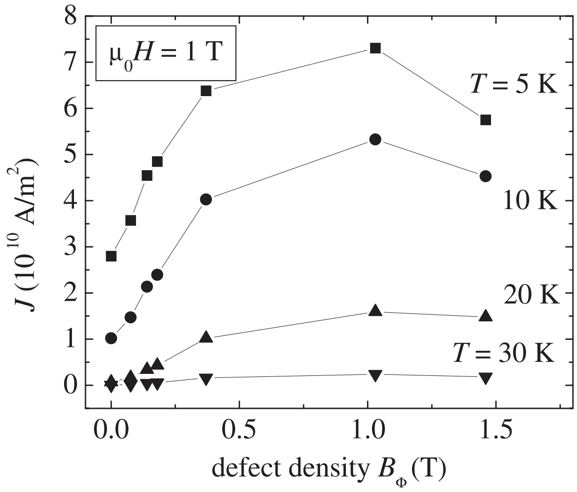

Figure 3. The persistent current density J against B_Φ, showing optimization of the defect density, for measurements in an applied magnetic field of 1 T at various temperatures.

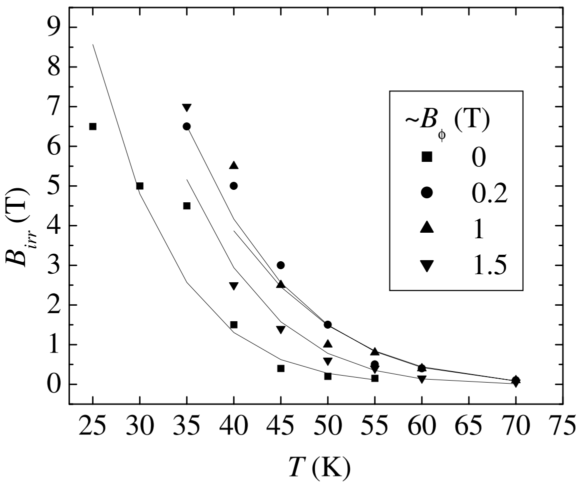

Figure 4. The irreversibility line $B_{irr}(T)$, at which the persistent current density becomes immeasurably small. The curves are power law fits to guide the eye.

At the highest fluence, the T_c is reduced by about 11 K from the value of 99 K observed for the virgin sample. The decrease is proportional to the proton fluence, showing that defect production is additive. The Meissner fraction $-M/H$ also decreases linearly with proton fluence (from 0.58 to 0.49), as is evident in figure 1(b). Both T_c and the Meissner fraction change by similar amounts, about 12–15%. Later we compare these modest changes with much larger changes in the mixed state equilibrium magnetization. First, however, we examine the impact of randomly oriented CDs on the irreversible properties of the Tl-2212 superconductor.

The addition of correlated disorder increases the persistent current density J in these materials, often by large factors. This is illustrated in figure 2, a plot of the intragranular current density at 30 K against magnetizing field, for various defect densities. The enhancement in J is modest at low fields, where naturally occurring defects in the virgin sample provide a modicum of pinning. With increasing field, however, J for the virgin superconductor falls rapidly and lies well below that for the processed materials; at intermediate fields, the artificial defects enhance J by nearly two orders of magnitude. The maximum J is attained at some optimum defect density, typically near $B_\Phi = 1$ T. For example, in figure 2, the data for the highest fluence, $B_\Phi = 1.5$ T, lie below those for $B_\Phi = 1$ T. This effect is seen more clearly in figure 3, which shows J as a function of B_Φ, measured at various temperatures in an applied field of 1 T. At higher defect densities, the deterioration of J may arise, at least in part, from the presence of additional CDs that promote the hopping of vortices to nearby empty pinning sites. Overall, the optimum defect density depends weakly on the temperature and applied field.

As the temperature and magnetic field increases, the effectiveness of vortex pinning deteriorates. Thus the persistent current density J decreases and at some field and temperature, it falls below the instrumental noise floor. This provides an experimental criterion for the irreversibility line, which here is $J_{criterion} = 500$ A cm^{-2} $= 5 \times 10^6$ A m^{-2}. The results for this experimental boundary $B_{irr}(T)$ are shown in figure 4 for several of the materials; in the figure, the full curves are power law fits to the data to serve as guides to the eye. Clearly, the addition of defects elevates the irreversibility line, by about 10 K for the case with $B_\Phi = 1.0$ T. In much of the phase field, however, a still higher defect density ($B_\Phi = 1.5$ T) shifts the irreversibility line backwards to lower temperatures, reducing the range of H, T space allowable for electric current conduction. In conclusion, the best performance for potential applications of these materials is achieved at relatively moderate irradiation doses ($B_\Phi \approx 1$ T).

Let us now consider the impact of adding randomly oriented CDs on the equilibrium magnetization in the mixed state. Overall, the fission induced columnar tracks produce a substantial reduction in the equilibrium magnetization M_{eq} and a systematic deviation from the 'standard' London field dependence behaviour in which M_{eq} is directly proportional to $(1/\lambda_{ab}^2)\ln(\beta B_{c2}/H)$, where λ_{ab} is the in-plane London penetration depth, β is a constant of order unity, and B_{c2} is the upper critical field [10]. To make visual the differences between materials, figure 5 shows M_{eq} as a function of applied field on a logarithmic axis. In this presentation, the isothermal

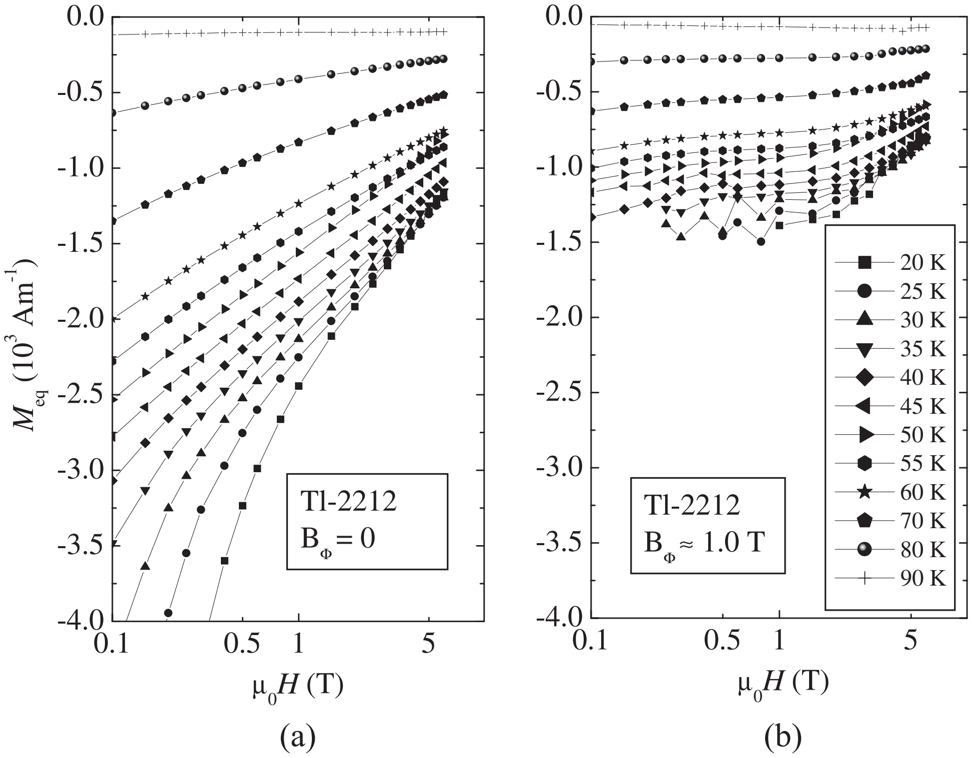

Figure 5. Experimental results for the equilibrium magnetization $M_{eq}(H, T)$ plotted against $\ln(H)$, for (a) virgin Tl-2212 and (b) a material with a defect density $B_\Phi = 1$ T. Note the large reduction in the magnitude of M_{eq} after irradiation.

data for the virgin material (figure 5(a)) are indeed linear, as predicted by London theory. (The deviations from linearity occur at low fields where H approaches H_{c1}, in violation of London theory, and at low temperatures where the materials become increasingly hysteretic.) From the slopes of the curves in figure 5(a) we can obtain values for $\lambda_{ab}(T)$, which are presented later.

The addition of fission-generated defects strongly modifies the mixed state magnetization. This is evident by examining the data for Tl-2212 containing defects with density $B_\Phi = 1.0$ T, which are shown in figure 5(b). Comparing the data for the virgin and irradiated samples, which are plotted with the same vertical scale, shows that the magnitude of M_{eq} decreased considerably, especially at a low flux density, with the addition of defects. Furthermore, the functional dependence on field changed considerably from the simple London dependence. Indeed, as seen clearly in figure 6, M_{eq} acquires an 'S'-like dependence on field. This 'anomalous' behaviour has been observed in cuprates containing parallel columnar defects formed by 5.8 GeV Pb ions, in thallium-based single crystals [11], in Bi-2223 tapes [12], and in Bi-2212 single crystals [13, 14].

The non-London behaviour of the equilibrium magnetization has been interpreted in terms of magnetic interactions between the vortex lattice and the CDs. Wahl *et al* [11] calculated the change in energy due to direct intervortex repulsion when a vortex is displaced from its natural position in the lattice to a particular CD. The work needed to deform the lattice must be compensated by the gain in pinning energy for occupation of the defect. Assuming parallel columnar tracks and a Poisson distribution of distances between tracks, Wahl *et al* [11] made

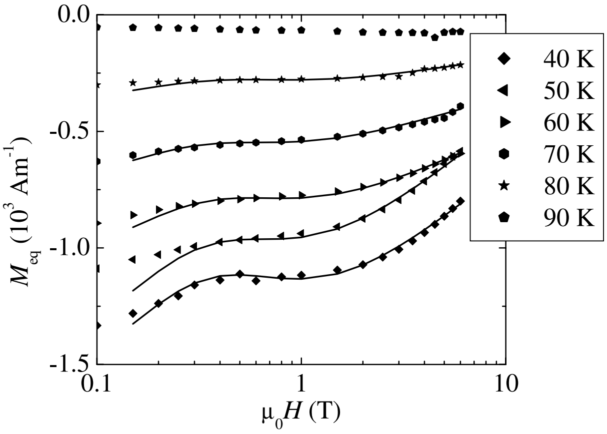

Figure 6. An expanded view of $M_{eq}(H, T)$ against $\ln(H)$ for Tl-2212 with $B_\Phi = 1$ T. The symbols are experimental results; the curves show the modelling of data using (1) which includes vortex–defect interactions.

a simple calculation of M_{eq} which explains the anomalous S-shaped behaviour observed in thallium-based single crystals containing parallel CDs.

In the present work, however, the fission-generated columnar microstructure is randomly oriented, which tends to entangle the vortices. Thus it is first surprising that the present system—polycrystalline with random CDs—behaves similarly to textured or single-crystal samples containing parallel defects. This phenomenon may arise, however, from an anisotropy-induced 'refocusing' of the defects and field toward the crystalline c-axis. Indeed, it is well known in studies of highly anisotropic single crystals that only the normal component of field is effective [15]. Studies on polycrystalline Hg-cuprates containing fission-generated random CDs showed [8] that increasing the mass anisotropy parameter γ (from Hg-1201 to Hg-1212 to Hg-1223) led to a recovery of vortex variable range hopping (VRH), very similar to that observed in YBaCuO single crystals containing parallel CDs [16]. With the high superconductive anisotropy of Tl-2212 ($\gamma > 100$), we can expect a corresponding 'refocusing' of the defect and vortex arrays. According to the theoretical development [8], the complexity of randomly oriented CDs in a polycrystalline material is reduced to some degree by a large superconducting anisotropy, restoring the simple physics of a crystal with parallel pins. Qualitatively, then, it is quite reasonable to attribute the changes in equilibrium magnetization to vortex pinning by random CDs.

Continuing this analysis, we next ask whether a reasonable quantitative description can be obtained, given the complexity of the materials in real space. Thus we use the vortex–defect interaction model of Wahl *et al* [11] which provides that

$$M_{eq} = -(\varepsilon_0/2\Phi_0) \times \ln(\eta H_{c2}/B) \\ -(U_0/\Phi_0)\{1 - [1 + U_0 B_\Phi/\varepsilon_0 B]\exp(-U_0 B_\Phi/\varepsilon_0 B)\} \quad (1)$$

where $\varepsilon_0 = (4\pi/\mu_0)[\Phi_0/4\pi\lambda_{ab}]^2$ is the line energy, U_0 is the pinning energy, and $B = \mu_0(H + M) \approx \mu_0 H$ since M is small. The first term is, of course, the conventional London expression; the second term represents a correction in intermediate fields that vanishes in large fields $B \gg B_\Phi$. Given the complex nature of the polycrystalline material and the vortex and defect arrays, we avoid any assumptions about temperature dependences or inter-relationships of the variables. Therefore we manually varied the free parameters in the above expression to achieve a reasonable representation of $M_{eq}(H)$ at each temperature. The resulting modelling, as shown as full curves in figure 6 for the sample with $B_\Phi = 1$ T, is reasonably successful; similar results were obtained for the sample with a higher defect density. The values used for B_Φ are reasonable, lying within 20% of those calculated from the proton fluence. Also, we have values for the pinning energy $U_0 = (0.9\text{–}1.2)\varepsilon_0$.

The remaining parameter of interest is the penetration depth. Results for λ_{ab} are shown in figure 7 as a plot of $1/\lambda_{ab}^2$ against temperature. For the virgin material, the values were obtained from a standard London analysis (the slopes of the curves in figure 5(a)). For the irradiated materials, however, the values were obtained from the rather empirical modelling described above. In fact, the results for all three cases are well behaved, with $1/\lambda_{ab}^2$ varying linearly with T near T_c, in keeping with Ginzburg–Landau theory, and extrapolating to zero near the T_c measured in low field. From the linear dependence on T, one can, in the spirit of the Ginzburg–Landau theory, extrapolate and estimate values of λ^2 at $T = 0$. (Since we are interested in the *relative* values of λ, uncertainties in the exact temperature dependence are not important, so long as the same linear extrapolation is used for the three samples.) A cursory examination of figure 7 shows that the proton irradiation increased λ significantly. Theoretically, Wahl *et al* [11] predicted that the addition of CDs should change the penetration depth, with

$$\lambda^{-2}(B_\Phi) = \lambda^{-2}(B_\Phi = 0) \times [1 - 2\pi R^2 B_\Phi/\Phi_0] \quad (2)$$

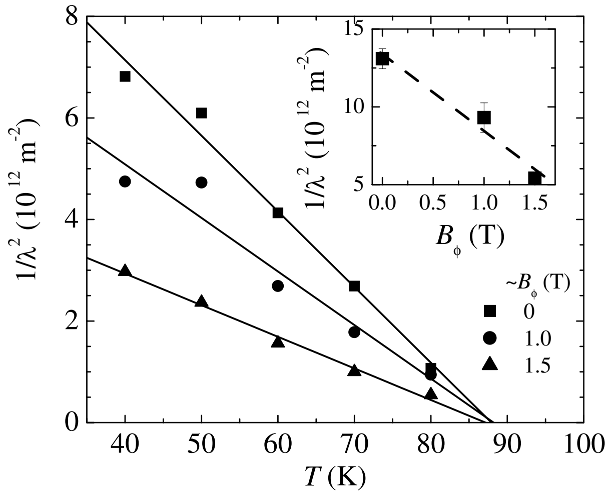

Figure 7. The London penetration depth against temperature T. For the irradiated materials, values come from modelling the equilibrium magnetization (see the text and figure 6); for the virgin sample, values come from a conventional London analysis of the data in figure 5. Inset: values of $1/\lambda^2(T = 0)$ plotted against B_Φ; the broken line shows the dependence given in equation (2).

where R is the radius of the columnar track. To test this dependence, the inset in figure 7 shows the extrapolated values of λ^{-2} plotted against B_Φ. As predicted, a roughly linear variation is observed, with a slope corresponding to $R \approx 11$ nm. This value is roughly two times larger than the transverse track dimension typically imaged by TEM. There are several factors that tend to increase this apparent value. One is the fact that the columnar tracks are not all perpendicular to the CuO planes; oblique tracks have a larger effective area. Second, Zhu and Suenaga have argued from TEM studies that columnar defects in YBaCuO have a 'halo' of damage extending to roughly twice the diameter of the high contrast, amorphized region. Third, conventional GLAG (Ginzburg–Landau–Abrikosov–Gorkov) theory predicts that the penetration depth λ^2 increases as $(1 + \xi_0/\ell)$ when the electronic mean free path, ℓ, is reduced by scattering [18]. This effect has been observed in $Bi_{1.8}Pb_{0.3}Sr_2Ca_2Cu_3O_{10}$ superconductors that were irradiated with fast neutrons to create point-like and collision cascade damage [19]. For the present study, we expect to have additional, fluence-dependent damage arising from neutrons and secondary protons released by spallation, immediately upon impact of a Tl nucleus by a 0.8 GeV proton. In total, each of these effects tends to increase λ and to produce larger values for the radius R.

4. Summary

Irradiation of polycrystalline $Tl_2Ba_2CaCu_2O_x$ superconducting materials with 0.8 GeV protons has produced significant increases in vortex pinning. The protons create randomly oriented CDs via a fission process in which recoil fragments amorphize the host superconductor. Enhancements in pinning increase the persistent current density observed magnetically and elevate the irreversibility line. For this material, a defect density corresponding to an effective matching field B_Φ

near 1 T produces optimum results. The addition of these defects markedly affects the equilibrium magnetization, reducing its magnitude significantly. In addition, its magnetic field dependence changes from a simple London $\ln(H)$ form (as observed for the virgin material) to a more complex dependence. By incorporating the influence of vortex–defect interactions and allowing for an anisotropy-induced 'refocused' vortex–defect array, we were able to model the field and temperature dependent magnetization, with reasonable parameter values. Thus, irradiation with highly penetrating gigaelectronvolt protons provides a mechanism for doping randomly oriented CDs deep within superconducting materials. This not only enhances the current-currying performance of the material, but also provides interesting physical systems with which to study the interaction of vortices with correlated disorder.

Acknowledgments

We thank M Paranthaman for providing the starting Tl-2212 materials used in this study. The work of JGO was partially financed by the Chilean FONDECYT, grant 1000394. Oak Ridge National Laboratory is managed by UT-Battelle, LLC for the US Department of Energy under contract DE-AC05-00OR22725. Los Alamos National Laboratory is funded by the US Department of Energy under contract W-7405-ENG-36.

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