FLUX PINNING IN NEUTRON IRRADIATED $YBa_2Cu_3O_{7-x}$

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Abstract

Identical polycrystalline samples of $YBa_2Cu_3O_{7-x}$ have been irradiated with fast neutrons (E > 0.1 Mev) in eight steps between 0 and 2.1×10^{18} n/cm^2. Notable irradiation effects include a T_c depression of nearly 2 K at the highest fluence and large improvements in the critical current density for fields from 0-9 T and temperatures between 4-80 K. Critical currents approaching 2×10^7 A/cm^2 are observed for optimally irradiated materials at 5K (in zero field) while at 77 K, J_c(0 kOe) approaches 5×10^5 A/cm^2. Irradiation is seen to take a nearly equilibrium magnetization curve at 77 K and broaden it to a significantly hysteretic curve. A substantial shift in the effective pinning potential as a function of current density is inferred from magnetic relaxation measurements at H = 1T. This is the first such measurement in which systematically increased activation energies for flux creep (as a function of current density) are noted.

Introduction

One of the most important issues pertinent to the application of high temperature superconductors concerns the development of a fundamental understanding of the way in which defect pinning may be altered so as to produce high J_c materials, especially at high temperatures. Recent experimental work clearly indicates that defects introduced by neutron irradiation in polycrystalline[1] and single-crystal[2] $YBa_2Cu_3O_{7-x}$ are effective in increasing the hysteretically measured J_c. The first of these studies[1], performed on a polycrystalline sample, indicated an approximately three-fold increase in the critical current density in zero field at 5K, with even more sizeable enhancements at high fields. The second study[2] on a single crystal sample revealed order of magnitude enhancements in J_c in high fields and at high temperatures. A hysteretically determined J_c of nearly 6×10^5 A/cm^2 was inferred for this crystal in a field of 0.9 T and at 77 K. Recent microstructural studies on irradiated and unirradiated samples (those of reference 1) have confirmed expectations that defects introduced by such radiation damage are homogeneously distributed and of a size comparable to the coherence length.[3] This work revealed strain induced contrast from defected regions 2-7 nm in size and comprising a 3% volume fraction in the irradiated sample.

Lacking in experiments to date is clear confirmation that the volume pinning energy is altered by irradiation. The issue of "giant" flux creep[4] and the inferences as to pinning energies which can be made from magnetic relaxation (or flux creep) measurements has been of notable importance in recent discussions of pinning. In previous studies of the influence of neutron irradiation on magnetic relaxation rates[5], larger activation energies were noted at 1T for a neutron irradiated Y123 sample as compared with an unirradiated control sample. These measurements, however, showed the apparent pinning potential to be a monotonically increasing function of temperature. This anomalous behavior has been explained recently as a manifestation of inherent non-linearity of the flux creep activation energy as a function of the current density, J, (or equivalently the magnetization)[6]. Recently, a technique for extracting the dependence of the flux creep activation energy[7] on current has been demonstrated. Further it is suggested that the so called $U_{eff}(J)$ functional dependence is the materials related parameter most appropriate for gaging the utility of the materials in current carrying applications. In what follows we describe experiments designed to further our understanding of the influence of neutron irradiation on critical current densities in the Y123 high temperature superconducting phase including the first determination of large shifts in the $U_{eff}(J)$ curve for a neutron irradiated sample.

Experimental Techniques

Polycrystalline samples of Y123, were produced following the procedure detailed in reference 1. Optical microscopy indicates an average grain size of approximately 10 μm for this material. The samples were determined to have nearly identical T_c's and hysteresis curves prior to irradiation. Subsequent irradiation was performed on eight samples yielding a set with fast neutron (E > 0.1 MeV) fluences of 0, 0.06, 0.24, 0.44, 0.79, 1.30, 1.87, and 2.12×10^{18} n/cm^2, respectively (including the unirradiated sample). Neutron irradiation was performed isothermally (80 °C) at the LANL Omega West reactor. Magnetic relaxation and hysteresis measurements were made using a PAR vibrating sample magnetometer equipped with a 9T superconducting solenoid. Hysteresis curves were obtained for fields between ± 9 T over a time period of ~45 minutes. Magnetic relaxation experiments in a field of 1 T were initiated on the virgin magnetization curve and were of ~1 hour duration.

Results and Discussion

Figure 1 illustrates the evolution of magnetic hysteresis with increasing fluence, at 5 K. Notable is the increasing width of the hysteresis curves with the largest incremental changes occurring at small fluences and saturating at the largest fluences. Initial magnetization curves follow a $B^{1/2}$ power law for fields below full flux penetration of the grains (and $H > H_{c1}$) and exhibit a $B^{-1/2}$ dependence for higher fields up to 9 T. The magnetic hysteresis is apparently just saturated for a fluence of 2.1×10^{18} n/cm^2 at 5 K. Similar enhancements were observed at temperatures up to 77 K, although the field dependences were not as simple and saturation was seen to occur at lower neutron fluences.

Similar hysteresis data at 60 K is shown in Figure 2a for fluences of 0, 0.06, 0.79, and 2.12×10^{18} n/cm^2, respectively. Here the saturation of the magnetic hysteresis was seen to occur at a fluence of 0.79×10^{18} n/cm^2. The figure shows nearly identical

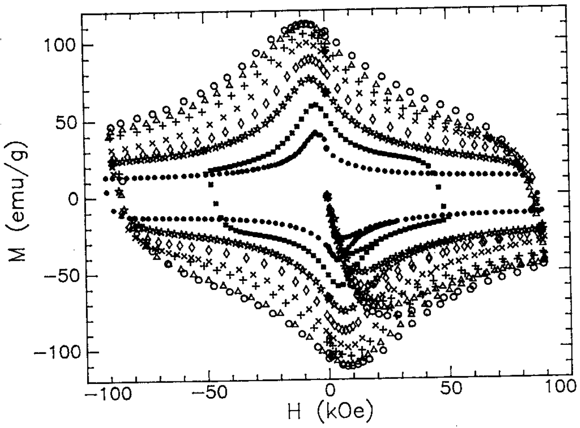

Figure 1: Magnetic hysteresis curves, at 5K, for Y123 samples having experienced neutron irradiation fluences of 0, 0.06, 0.24, 0.44, 0.79, 1.30, 1.87, and 2.12×10^{18} n/cm^2, respectively. Hysteresis increases with fluence over this range.

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hysteresis for the 0.79 and 2.1 x 10^{18} n/cm^2 irradiated samples. Virtually no differences in hysteresis are observed for any of the fluences between these values. In several respects this data is more interesting than the 5 K hysteresis. First, it can be seen that the unirradiated sample displays a "butterfly" shaped hysteresis loop implying a slight peak effect in the $J_c(H)$ dependence. Similar effects have been observed by Larbalestier and Daeumling[8] (on a much larger ΔM scale) and attributed to the fact that oxygen deficient regions in the superconductor can be "turned on" as pinning sites above the field or temperature where they are driven normal. Larbalastier and Daeumling also showed that eventually these same defects lead to granularity at still higher temperatures or fields where these defects become multiply connected (i. e the percolative limit). This notion is supported by our observations of the magnetic hysteresis of the virgin sample at 77 K, as shown in Figure 2b, where much of this butterfly effect has disappeared. Even more notable are the extremely large increases in magnetic hysteresis as a function of neutron fluence, exhibited at 77 K. The figure clearly shows enhancements of a factor of ~10 in the hysteresis in certain field ranges for the optimally damaged material. The butterfly shape appearance eventually disappears as a function of increasing fluence suggesting that the neutron induced defect structure is significantly more effective in pinning flux than the proposed oxygen deficient regions. This may imply that the size scale of the neutron induced damage may be much finer than that of the defected regions responsible for the butterfly effect. It should be noted that it is conceivable that the neutron induced damage itself is largely related to disorder on the oxygen sublattice.[3]

The fact that the increases in hysteresis eventually saturate as a function of neutron fluence is suggestive. We hypothesize that the effects of the increased pinning site density introduced by the neutron irradiation and the depression of T_c act to counterbalance one another at high temperatures so that the fluence at which saturation of the increasing hysteresis occurs is strongly temperature dependent. Figure 2b clearly supports this claim. For the 77 K data saturation with respect to fluence occurs at a fluence of 0.44 x 10^{18} n/cm^2. Again little difference in the hysteresis was apparent beyond this fluence. The 77K data for the virgin sample shows the most nearly reversible hysteresis of any depicted. The influence of neutron irradiation is extremely pronounced in the low doses leading to nearly 20-fold increases in the magnetic hysteresis in certain field ranges. Perhaps the most striking result is the fact that the magnetic irreversibility line is clearly shifted to higher fields in the first increments of irradiation at 77K.

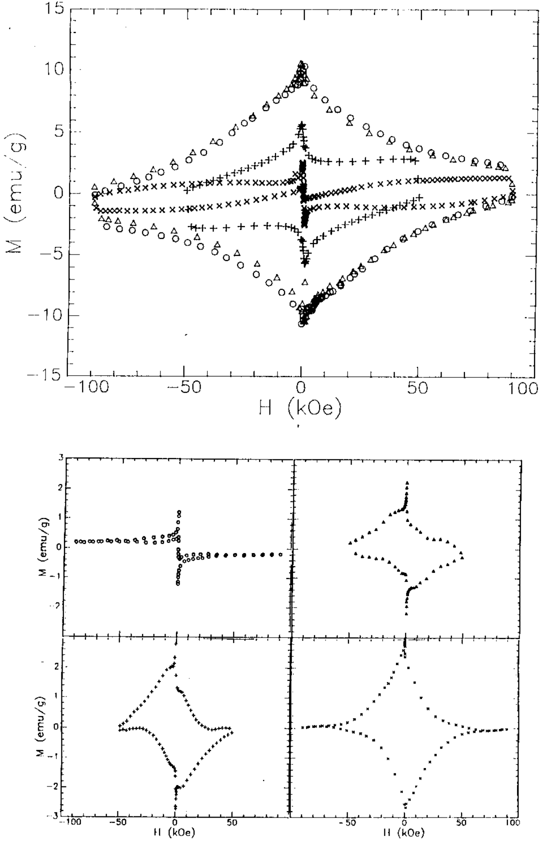

Figure 2: Magnetic hysteresis curves for Y123 samples at (a) 60 K having experienced fluences of 0, 0.06, 0.79, and 2.12 x 10^{18} n/cm^2, respectively and (b) 77 K having experienced fluences of 0, 0.06, 0.44, and 2.12 x 10^{18} n/cm^2 (as shown in the upper left upper right, lower left and lower right,respectively).

Critical current densities derived from the hysteresis data using the Bean[9] model for a slab geometry of average size ~10 μm. are shown in Figures 4a and 4b for 5 K and 77 K respectively. Note that the 5 K data exhibits a zero field Jc which is just saturated for the highest fluences while the 77 K data shows Jc saturation by a fluence of 1 x 10^{18} n/cm^2. A zero field critical current density of 1.9 x 10^7 A/cm^2 is apparent for the neutron irradiated sample at 5 K while 0.7 x 10^7 A/cm^2 is observed in the unirradiated sample. A nearly 3-fold increase in the zero field J_c at low temperatures is consistent with earlier measurements on similar neutron irradiated samples[1]. At 77 K, a zero field J_c of 4.5 x 10^5 A/cm^2 is observed in the irradiated sample increased from ~ 2 x 10^4 A/cm^2 for the

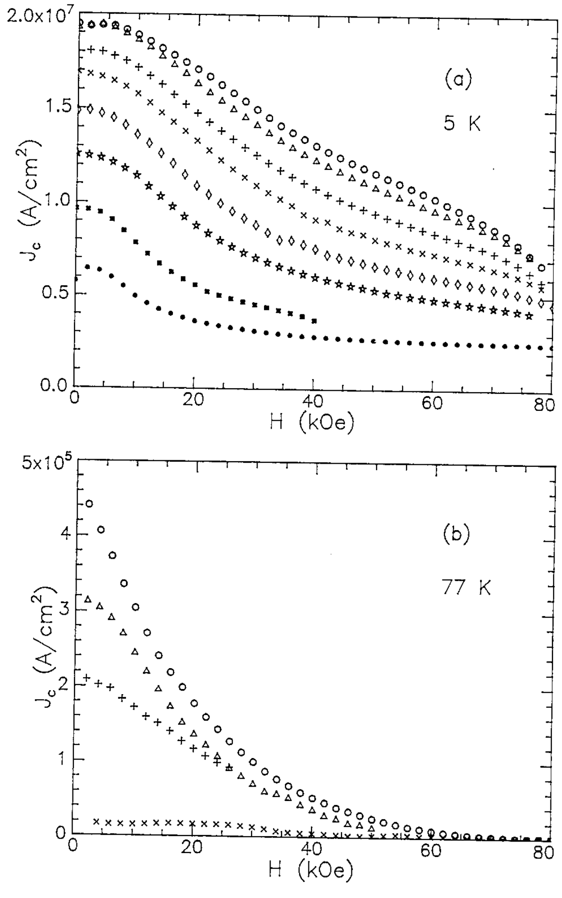

Figure 3: Critical current densities for Y123 as a function of fluence as determined from the magnetic hysteresis using the Bean model expression $J_c = 15\Delta M/d$ where ΔM is the magnetic hysteresis in emu/cm^3 and d is the grain diameter taken as 10 μm. (a) J_c (H) at 5 K for neutron irradiation fluences of 0.06, 0.24, 0.44, 0.79, 1.30, 1.87, and 2.12 x 10^{18} n/cm^2 and (b) J_c (H) at 77 K for neutron irradiation fluences of 0, 0.06, 0.24, and 2.12 x 10^{18} n/cm^2.

unirradiated sample. This 20-fold at 77 K increase is consistent with the observation on a single-crystal of Y123[2]. Further, J_c(0.9 kOe) is 4.0×10^5 A/cm^2 at 77 K for the irradiated sample similar to the reported 6×10^5 A/cm^2 for a single crystal.

The critical current densities shown in Figure 4 should be viewed with the well known inadequacies of the Bean model expression in mind. In particular the critical current density in low fields tend to by underestimated. It should be further recognized that J_c's determined hysteretically are reduced from their critical state values by the creep which has occurred on the experimental time scale. In fact, hysteretically determined $J_c(T)$ data at fixed fields is observed to obey an $\exp(-T/T_0)$ the significance of which will be explored in a subsequent publication.

Magnetic relaxation data is now commonly used to infer volume pinning energies in high temperature superconducting materials. The often used method for analyzing this data has been to invoke a linear approximation for the effective pinning potential (i.e. the volume pinning energy reduced by a Lorentz force related energy) given by the expression:

$$U_{eff} = U_0 - JBVa \qquad (1)$$

where J represents the magnetization current, B is the local flux density, V is a correlated volume for flux motion and a is a hop distance. This linear model used in the flux creep formalism allows for the determination of U_0 from magnetic relaxation measurements through determination of the logarithmic magnetic relaxation rate, dM/dln(t). Recently, some problems with the use of this linear model have been delineated.[5-7] First, activation energies, U_0's, derived from magnetic relaxation measurements at high J^4 (~J_c) are generally significantly smaller than those derived from thermally activated resistance measurements at low J (<< J_c).[10] Second, U_0's, derived from magnetic relaxation studies are observed to increase monotonically with temperature over a broad range. [5,6]

It has recently been suggested that the shape of a non-linear $U_{eff}(J)$ potential can be probed using an analysis which appeals directly to the rate equation[7]:

$$\frac{dM}{dt} = \frac{H\nu a}{2\pi d}\exp\left(\frac{-U_{eff}}{kT}\right) \qquad (2)$$

where ν is the flux hopping attempt frequency, H is the applied field and d is the grain size. This rate equation is directly obtainable from that of Beasley et al.[11] in a 1-d model in a slab geometry after integration to arrive at a an expression in terms of the average B or magnetization which are measured quantities in flux creep experiments. Rearranging expression (2):

$$\frac{U_{eff}}{k} = -T\ln\left|\frac{dM}{dt}\right| + T\ln\left(\frac{H\nu a}{2\pi d}\right) \qquad (3)$$

yields a prescription for obtaining the effective pinning potential from M(t) data recognizing that the argument of the second logarithm is essentially constant for a fixed field and T << T_c where temperature scaling laws[12] pinning energies are relatively flat.

Magnetic relaxation measurements have been made on the unirradiated sample and the irradiated sample with a fluence of 2.1×10^{18} n/cm^2. These measurements have been made in an applied field of 1 T after cooling to the chosen temperature (i.e relaxation from the virgin magnetization curve). The time dependence of the magnetization was then monitored over ~ 1 hour periods. The derivative of the magnetization with respect to time is estimated by ΔM/Δt over the short interval between successive magnetization points. For relaxation at different temperature the magnetization values decrease with increasing temperature allowing for probing of the thermally activated response over a range of magnetizations and consequently current densities. In actuality it is only the irreversible magnetization which is of interest, however for the temperatures reported here, the equilibrium magnetization has been determined to be a negligibly small portion of the total magnetization at H = 1T. The relaxation data is transformed into the form required by (3) in the manner described in reference 7. Figure 4 shows the M (~J) dependence of U_{eff} derived from magnetic relaxation measurements (performed between 5 and 40K) for the unirradiated sample and the irradiated sample. In the case of the irradiated sample using $\ln\frac{H\nu a}{2\pi d} = 24$ yields a $\frac{U_{eff}}{k}$ behavior which is relatively smoothly varying as a function of M or equivalently J. For the unirradiated sample a value of 18 was employed.

Notable in the data of Figure 4 is the extraordinary increase in the effective potential for the irradiated sample as compared (at similar values of J) with the unirradiated sample. One also notes that U_e values are similar for the unirradiated and irradiated sample at a given temperature. This is a natural consequence of the thermally activated flux penetration process. A change in the energy scale for pinning is consistent with the observed higher critical current densities (though not necessary to explain them) as well as shifts in the irreversibility line (which do seemingly require increased pinning energies). These observations can be taken as evidence that pinning in Y123 is not limited by intrinsic pinning considerations i.e. defect pinning is a viable means of increasing J_c and more notably the temperature dependence of J_c insofar as it is limited by flux creep. Further, this data is well fit by a ln(J) functional dependence consistent with that seen in previous studies of grain aligned Y123[7] and also in transport measurements.[13] The logarithmic dependence of U_{eff} is reconcilable with an observed exponential dependence of the hysteretically determined J_c on temperature whereas the simple linearized flux creep model predicts a linear temperature dependence which is clearly inadequate in explaining our data. We have made similar observations of exponentially varying J_c's with temperature for a $La_{1.86}Sr_{0.14}CuO_4$ single crystal. These results and the reconciliation of such a temperature dependence with a non-linear effective potential will be explored in a forthcoming publication. Although a microscopic mechanism in which a particular (logarithmic) spatial dependence of the pinning well has been proposed by Zeldov et al.[13] we do not rule out the possibility that the ln(J) dependence is consistent with a distribution of pinning energies as proposed by Hagen and Griessen[14] or by a crossover between two or more pinning regimes describable by power law fits of $U_e(J)$ such as those suggested by Feigelman et al.[15].

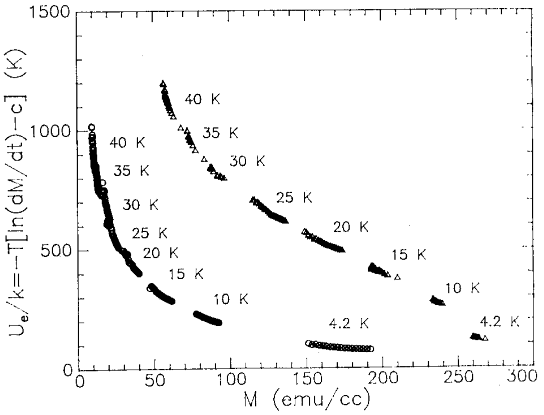

Figure 4: Plots of magnetic relaxation data, $T\,(\ln\frac{dM}{dt} + C)$, where the constant C has been determined to be 18 for the unirradiated sample and 24 for the irradiated sample subtracted from each data set to yield U_{eff} (in units of absolute temperature) as a function of the magnetization for unirradiated and irradiated Y123. Curves reflect separate magnetic relaxation experiments between 4.2 and 40 K as labelled.

Conclusions

Irradiation, with fast (E> 0.1 MeV) neutrons of a polycrystalline Y123 sample has been shown to dramatically influence the magnetic hysteresis and hysteretically derived J_c. Most notable are the enhancements at high temperatures. The irreversibility line is shown to be shifting at 77 K with increasing neutron fluence. The fluence at which beneficial enhancement in hysteresis and critical current densities saturates is seen to be strongly dependent on temperature.

For the first time it has been shown that the improved J_c is accompanied by concomitant increases in the effective pinning potential . This result carries the obvious implication that flux creep related problems have the significant possibility of reduction if suitable defects can be introduced as is the case for neutron irradiation. This perhaps leads to a more optimistic outlook for the eventual application of these materials.

References

1. J. O Willis, J. R. Cost, R. D. Brown, J. D. Thompson, and D. E. Peterson; Mat. Res. Soc. Proc. **99**, 391, (1988)

2. R. B. VanDover, E. M. Gyorgy, L. F. Schneemeyer, J. W. Mitchell, K. V. Rao, R. Puzniak, and J. V. Waszczak, Nature **342**, 55, (1989). The first neutron irradiation study on a single crystal of Y123 was reported by: A. Umezawa, G. W. Crabtree, J. Z. Liu, H. W. Weber, W. K. Kwok, L. H. Nunez, T. J. Moran, C. H. Sowers, and H. Claus, Phys. Rev. B **36**, 7157, (1987). The later VanDover et al. work reports extraordinarily large enhancements in J_c especially in field.

3. J. -W. Lee, H. S. Lessure, D. E. Laughlin, M. E. McHenry, S. G. Sankar, J. R. Cost, J. O. Willis, and M. P. Maley, accepted Appl. Phys. Lett.

4. Y. Yeshurun and A. P. Malezemoff, Phys. Rev. Lett.. **60**, 2202, (1988).

5. M. E. McHenry, J. O. Willis, M. P. Maley, J. D. Thompson, J. R. Cost and D. E.. Peterson; Physica C **162-164**, 689, (1989).

6. Y. Xu, M. Suenaga, A. R. Moodenbaugh, D. O. Welch, Phys. Rev. B **40**, 10882, 1989.

7. M. P. Maley, J. O. Willis, H. Lessure, and M. E. McHenry, Phys. Rev. B **42**, 2639, (1990).

8. M. Daeumling, J. M. Seuntjens and D. C. Larbalastier, Nature, **346**, 332 (1990); D. C. Larbalastier and M. Daeumling, preprint.

9. C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962).

10. T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, J. V. Waszczak, Phys. Rev. Lett. **61**, 1662, (1988).

11. M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682, (1969).

12. M. Tinkham, Phys. Rev. Lett. **61**, 1658, (1988)

13. E. Zeldov, M. M. Amer, G. Koren, A. Gupta, M. W. McElfresh and R. J. Gambino, Appl. Phys. Lett. **56**, 680, (1990).

14. C. W. Hagen and R. Griessen, Phys. Rev. Lett. **62**, 2856, (1989).

15. M. V. Feigelman, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Phys. Rev. Lett. **63**, 2303, (1989).