

EFFECTS OF NEUTRON IRRADIATION ON THE LONDON PENETRATION DEPTH FOR POLYCRYSTALLINE $\text{Bi}_{1.8}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ SUPERCONDUCTOR.

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ABSTRACT

Magnetization studies of polycrystalline $\text{Bi}_{1.8}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ superconductor, prior to and after neutron irradiation, showed an increase in J_c due to irradiation damage. Analysis of the *equilibrium* magnetization revealed significant increases in other more fundamental properties. In particular, the London penetration depth increased by $\sim 15\%$ following irradiation with 8×10^{16} neutrons/cm². Corresponding changes were observed in the upper critical magnetic field H_{c2} . However, the most *fundamental* thermodynamic property, the superconductive condensation energy F_c , was unaffected by the moderate level of neutron-induced damage.

INTRODUCTION

Magnetometric studies have been widely used to determine the physical properties of high temperature superconductors (HTSC).¹ For example, the intragrain persistent current density $J_p(T,H)$ in the mixed state can be found from measurements of the *irreversible* magnetization by means of the Bean model, provided the grain morphology is known. The London penetration depth λ can often be obtained from the logarithmic slope of the equilibrium (*reversible*) magnetization $M(H,T)$ using the standard London-limit formalism of Kogan et al.² More recent theoretical developments, however (Bulaevskii et al.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 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 provides a useful tool to determine the superconducting volume fraction of the material, as well as the characteristic magnetic fields and lengths. Cho *et al.*⁵ showed that this formalism still applies to random polycrystals, provided 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-Clem⁶ provides an alternative formulation for determining the fundamental properties of the superconductor from studies of the reversible magnetization in the intermediate field region.⁷

In this work, the main objective is to investigate the impact of neutron-induced damage on the London penetration depth. Here we used fast neutron irradiation to modify the Bi-based superconductor $\text{Bi}_{1.8}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$. Apparently the localized damage affects the superconductor in two ways: (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 providing vortex pinning sites. Here we study primarily mean free path effects, as there is little information to date regarding their impact on high- T_c superconductors.

EXPERIMENTAL ASPECTS

The sample was a polycrystalline specimen of the $\text{Bi}_{1.8}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ (hereafter BiPb-2223) compound, containing three adjacent CuO-layers in the unit cell. This highly anisotropic superconductor initially had a diamagnetic onset transition temperature $T_c \approx 109$ K, measured at low magnetic field ($\leq 4 - 10$ Oe). It consisted of thin platelets with edge dimensions of ≈ 100 microns. Two pieces of the same batch were used for comparison; one of them ($m = 169$ milligrams) was irradiated with $\sim 8 \times 10^{16}$ [n/cm^2] fluence of fast neutrons ($E > 0.1$ MeV), giving roughly a 3-fold increase of irreversible magnetization $M(H, T)$, i.e., a 3-fold increase in the critical current density J_c . The other piece ($m = 85$ milligrams) was left unirradiated. Both showed clear evidence of vortex fluctuation effects at temperatures of a few degrees below and above the low-field T_c .

The magnetic measurements were conducted using a multipurpose SQUID magnetometer. Both the virgin and the irradiated samples were measured in identical conditions, using a magnetometer scan length of $2 \sim 3$ cm, and fixed temperature settings ranging from 40 K to 108 K. To correct for background signals, the normal state magnetic susceptibility was measured for temperatures up to 250 K; the background moment was practically independent of temperature. For most of the study in the superconducting state, the measurements were conducted in the reversible region above the irreversibility line in the magnetic phase diagram.

The equilibrium magnetization $M(T,H)$ of the sample was calculated as the reversible magnetic moment per unit volume of BiPb-2223 compound. Normally, 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/cm³]). This procedure assumes that all of the BiPb-2223 material is superconducting. The determination of the *superconducting* volume of the sample, however, is a difficult step. In this work, we obtain the fraction of the total volume (the volume ratio " ϵ " = $V_{\text{SC}}/V_{\text{tot}}$) that is superconducting, from the fluctuation analysis. Using the procedure described previously,^{4,8} we obtain the results that ϵ was ~ 0.31 for the virgin sample and ~ 0.36 for the irradiated one.

THEORETICAL FRAMEWORK

According to Bulaevskii et al.³ and Kogan et al.⁴, fluctuations introduce a correction term, due to entropic effects, into the standard London-limit result $M \propto \ln(H)$, where M is the mixed state reversible magnetization. With fluctuations included, one has

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

where

$$g(T) = 32\pi^2 k_B T \lambda_{ab}^2(T) / (\phi_o^2 s) \quad (2)$$

The quantity M_o is the magnetization of a single crystal with $H \perp$ (layers); λ_{ab} is the magnetic penetration depth corresponding to screening by supercurrents in the ab planes; ϕ_o is the flux quantum; and s is the interlayer spacing. For BiPb-2223 containing three adjacent trilayers, we take s to be the separation of trilayers sets, 1.8 nm. At the characteristic temperature $T^* < T_c$ where $g(T^*) = 1$, M is independent of field and has the value $M^* = k_B T^* / \phi_o s = m^* / V_{\text{sc}}$. The field-independence of the mixed state magnetization at temperature T^* is a very important signature of the vortex fluctuation theory as well as the nonperturbative scaling theory.^{9,10,11} This relation also provides a means for calculating the superconducting volume V_{sc} and fraction ϵ , since the other quantities are measured. The results are valid for temperatures near and below T^* , where the correction term markedly affects the deduced values of $\lambda_{ab}(T)$. The above theoretical expressions refer to the case with $H \perp$ (layers), as with a single crystal. According to Cho et al.,⁵ these results still apply for a random polycrystal, if the material is sufficiently anisotropic that only the component of H perpendicular to the layers is effective. Then the magnetization M_s of a polycrystal is

$$M_s = M_o \langle \cos \theta \rangle + \langle \cos \theta \ln(\cos \theta) \rangle \frac{\phi_o}{32\pi^2 \lambda_{ab}^2} (1-g) \quad (3)$$

The brackets $\langle \dots \rangle$ denote an angular average with weighting factor $\sin \theta$. For random crystallites, one has $\langle \cos \theta \rangle = 1/2$; $\langle \cos \theta \ln \cos \theta \rangle = -1/4$; $M_s^* = (1/2)M_o^*$; and $(\partial M_s / \partial \ln H) = (1/2)(\partial M_o / \partial \ln H)$. In this work we have used equations 1-3 and the weighted angular averages to obtain λ_{ab} and related properties.

For comparison, the Hao-Clem⁶ analysis was used to compute the Ginzburg-Landau 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 G-L expressions. The procedure, which is not simple, has been described in other publications.⁷ A third theoretical model used in this work was the two dimensional scaling theory.^{9,10,11} Details of these analyses will be presented elsewhere.

RESULTS AND DISCUSSION

At elevated temperatures, the theory outlined above predicts that the magnetization should vary linearly with $\ln(H)$. This feature is clearly seen in Fig. 1, a semilogarithmic plot of M_s vs H for the unirradiated sample. The results for the neutron-irradiated specimen were very similar.

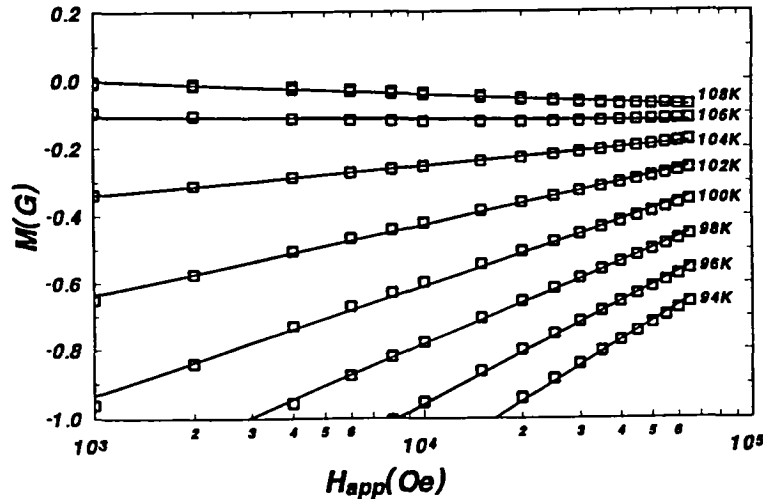


FIG. 1. Magnetization M_s versus H_{app} on a semilogarithmic scale, for non-irradiated (virgin), polycrystalline BiPb-2223 superconductor. Straight lines show the theoretical dependence at elevated temperatures.

Another feature evident in Fig. 1 is the horizontal, i.e., field independent magnetization for a temperature near 106 K. This locates experimentally T^* with the corresponding value M_s^* . From this and the relations cited, we obtained values for the superconducting volume fraction ϵ given above.

To determine the penetration depth $\lambda_{ab}(T)$, we obtain logarithmic slopes $\partial M_s / \partial \ln H$ from the data in Fig. 1 and similar

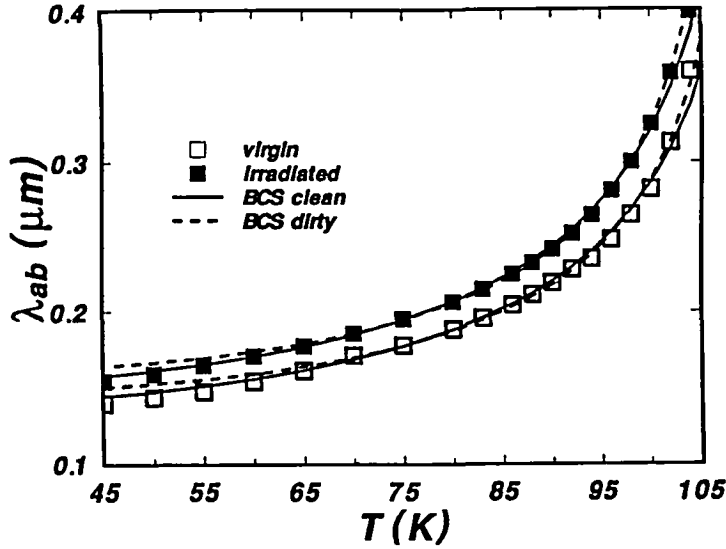


FIG. 2. $\lambda_{ab}(T)$ as a function of T from fluctuation analysis, for virgin and neutron irradiated BiPb-2223 materials. Dashed and solid lines are BCS fits to the data in the clean and dirty limits, respectively.

both limiting cases. In the more appropriate clean limit, the respective values are 139 nm and 113.8 K for the unirradiated sample, compared with 152 nm and 112.9 K for the irradiated material. Observe that in each case, $T_{co,\lambda}$ lies several Kelvins *above* the low field diamagnetic onset T_c values, 109 and 108 K, respectively. This is another characteristic feature of the vortex fluctuation theory.

A central result of this study is the increase in London penetration depth λ upon irradiation. Qualitatively, we attribute this feature to increased "dirtiness," i.e., disorder in the material, which scatters the conduction electrons. Another feature commonly associated with electron scattering and the associated reduction on electronic mean free path is an increase in the slope of the upper critical magnetic field $H_{c2}(T)$. Further analysis using the Bulaevskii-Kogan formalism indeed revealed a substantial steepening of the slope, by approximately 25-40 %. This increase was corroborated by complementary analyses using the Hao-Clem and scaling methods. An important consistency check is provided by the thermodynamic critical field $H_c(0)$. This was calculated both from the Hao-Clem formalism and from $\lambda_{ab}(0)$ and the coherence length $\xi_{ab}(0)$. We obtain nearly the same value, $H_c(0) \approx 1.3$ T, for both the virgin and the irradiated samples. This result is consistent with the expectation that the superconductive condensation energy $F_c = H_c^2/8\pi$ should not be affected by moderate densities of neutron generated defects. Details will be presented elsewhere in a comprehensive discussion.

plots. Analysis using Eq. 1 and 2 with appropriate angular and volumetric factors " ϵ " leads directly to λ_{ab} for each isotherm. The results for the BiPb-2223 virgin and irradiated materials are summarized in Fig. 2. The solid and dashed lines are BCS temperature dependencies in the clean and dirty limits, respectively, which were fitted to the data. Points for $T > T^*$ (outside of the range of validity of the theory) were not fitted. From the BCS fits, the extrapolated $\lambda_{ab}(0)$ and the *mean field* transition temperature $T_{co,\lambda}$ (where λ diverges) are derived in

CONCLUSION

This work shows that neutron-generated damage increased the London penetration depth of BiPb-2223 superconductor by approximately 10 ~ 15 %. Further analysis using three independent theoretical models has shown that $H_{c2}(0)$ concurrently increased by 25% ~ 40%. However, neither the thermodynamic critical field H_c nor the transition temperature T_c were appreciably affected. The observed behavior is similar in magnitude and directions to observations¹² in A-15 superconductors. For instance, Nb₃Sn and V₃Si thin films with differing residual resistivities just above T_c showed a significant increase in $H_{c2}(0)$ (about \approx 35 %) as the material goes from the clean (low resistivity) limit to the dirty (high resistivity) limit at the expense of a small decrease in T_c . The similarity of behavior suggests that the observed changes for the BiPb-2223 originate from mean free path effects. Further investigations are under way to test this hypothesis.

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