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# Large critical current density in neutron-irradiated polycrystalline $HgBa_2CuO_{4+\delta}$

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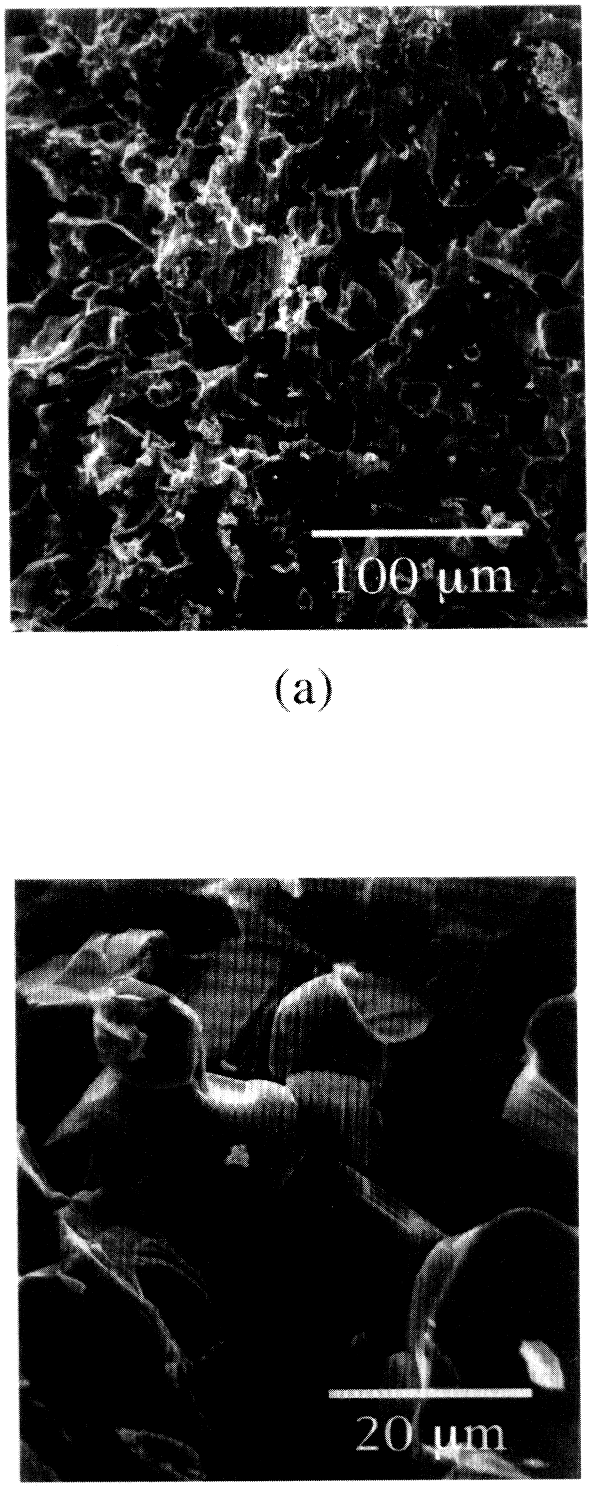
The effects of neutron irradiation on the superconducting properties of $HgBa_2CuO_{4+\delta}$ are reported. Enhancements in the magnetization critical current density ($J_c$), as calculated by the Bean model, vary with magnetic field and temperature and range from 1–2 orders of magnitude. Large values ($J_c > 10^4$ A/cm$^2$) are obtained above (0.5 T, 77 K) and (3 T, 50 K) after irradiation. A large shift in the irreversibility line toward higher field and temperature is also reported. These results demonstrate that $HgBa_2CuO_{4+\delta}$ is not intrinsically limited to low current density and may become a technologically important compound.

The recent discovery[1] of superconductivity at 94 K in $HgBa_2CuO_{4+\delta}$ is particularly noteworthy due to the high critical temperature ($T_c$) in a material containing only a single metal oxide layer. Owing to the resulting close spacing between $CuO_2$ planes, the superconducting properties in a magnetic field may be superior to other known oxide superconductors.[2] The first reports of the magnetic behavior[3,4] indicate that the irreversibility line is significantly higher than the two-layer compound $Bi_2Sr_2CaCu_2O_8$ and the three-layer compound $Bi_2Sr_2Ca_2Cu_3O_{10}$ , but below that of oxygen deficient $YBa_2Cu_3O_{7-\delta}$ and $TlBa_2Ca_2Cu_3O_9$. The reported critical current densities were rather low for all but the lowest temperature.[4] One demonstrated approach to improve the magnetic behavior is to introduce defects by irradiation—both neutron and ion irradiation can be effective.[5–10] Here we report measurements of large critical current densities and a significantly enhanced irreversibility line in $HgBa_2CuO_{4+\delta}$ by neutron irradiation. As technologically relevant current density is achieved at high temperature in a magnetic field, these results demonstrate that $HgBa_2CuO_{4+\delta}$ is not intrinsically limited to low current density and may become a technologically important compound.

Superconducting samples were formed from stoichiometric mixtures of HgO, $BaCO_3$, and CuO as described previously.[11] X-ray diffraction data using a rotating anode source indicated the sample to be single-phase $HgBa_2CuO_{4+\delta}$ without detectable impurities. This was confirmed by scanning electron microscopy (SEM) using a Hitachi S-800 equipped with a Link energy dispersive spectrometer (see Fig. 1), which showed a nearly phase-pure material with significant porosity and a layered structure within grains. ac susceptibility in a 1 gauss field at 100 Hz using a Lake Shore Cryotronics susceptometer exhibited a sharp transition (onset $T_c = 95.5$ K, $50\% T_c = 92.5$ K) with 100% shielding. Lastly, the polycrystalline rodlike samples ($4 \times 1.5 \times 1$ mm$^3$) were vacuum sealed in quartz tubes.

Superconducting properties were measured in a Quantum Design dc superconducting quantum interference device (SQUID) before and after neutron irradiation. The critical temperature was measured while warming in a 50 gauss magnetic field after zero-field cooling. Magnetic hysteresis loops were measured for magnetic fields ranging between ±5.5 T at 5, 20, 30, 40, 50, 60, and 77 K. Neutron irradiation was performed at the University of Illinois Advanced TRIGA reactor facility as described



(a)

(b)

FIG. 1. Scanning electron micrographs of $HgBa_2CuO_{4+\delta}$ with magnification of (a) 300× and (b) 1500×, illustrating a nearly phase pure material with significant porosity and a layered structure within grains.

previously[5,6] with sample temperature held below 100 °C to minimize annealing.[10] The 2.5 h irradiation corresponds to a thermal fluence of $2\times10^{17}$ neutrons/cm$^2$ and a fast fluence ($E > 0.1$ MeV) of $2.4\times10^{17}$ neutrons/cm$^2$.

The irradiation effect on $T_c$ was minimal—a uniform decrease of 1 K in the onset. The changes in the magnetic hysteresis, however, were dramatic. Figure 2 shows the magnetization hysteresis at 20 and 40 K after irradiation and at 20 K before irradiation. Both the magnitude and shape of the loops are changed. Before irradiation the hysteresis is comparable to the reversible magnetization, giving magnetization loops of one sign at 20 K. After irradiation, the hysteresis dominates the reversible magnetization and the loops are nearly symmetric about $M = 0$, as is typical in strongly pinned superconductors.

Critical current densities have been extracted from the hysteresis loops using a modified form of Bean's critical-state model.[12] Thus, $J_c = 20\Delta M/[f(1-f/3g)]$ has been used, where $\Delta M$ is the width of the hysteresis loop and $f$ and $g$ are face dimensions of a rectangular superconducting sample. As it is difficult to assess the coupling between superconducting grains and thus to determine values for $f$ and $g$ that correspond to the actual current flow paths, two critical current densities are reported: a high value derived from the average size of individual superconducting grains as observed by SEM ($f = g = 10$ µm), and a low value determined by considering the entire sample as the critical geometry ($f - 1$ mm, $g = 1.5$ mm). The high value represents separate current loops within individual grains, the low value represents current loops on the scale of the entire sample. Considering the sample porosity (see Fig. 1), however, we consider the high value to be indicative of the material properties, independent of the granularity of the material.

Figures 3(a) and 3(b) show the resultant $J_c$ as a function of magnetic field and temperature before and after irradiation, respectively. Note that the minimum value of the left ordinate has been fixed at $10^4$ A/cm$^2$—a minimum but not necessarily sufficient value for technological relevance.[13] Before irradiation, significant $J_c$ exists only at 5 and 20 K. After irradiation, however, relevant $J_c$ is obtained over the entire range of measured

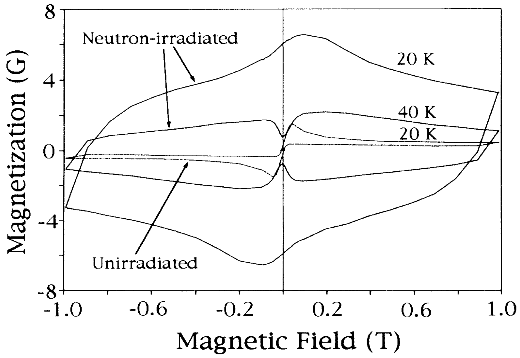


FIG. 2. Magnetization hysteresis loops at 20 and 40 K after neutron irradiation and at 20 K before irradiation as measured by a SQUID magnetometer.

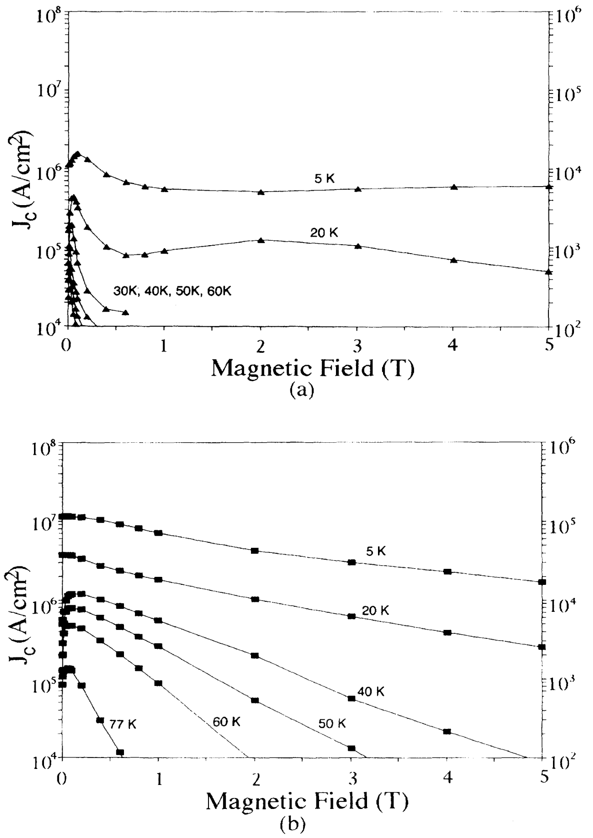


FIG. 3. Critical current density as a function of magnetic field and temperature (a) before and (b) after irradiation as derived from magnetization hysteresis measurements and Bean's critical state model. The high values (left ordinate) are based upon zero grain coupling and the low values (right ordinate) are based upon perfect grain coupling within the polycrystalline rod of $HgBa_2CuO_{4+\delta}$.

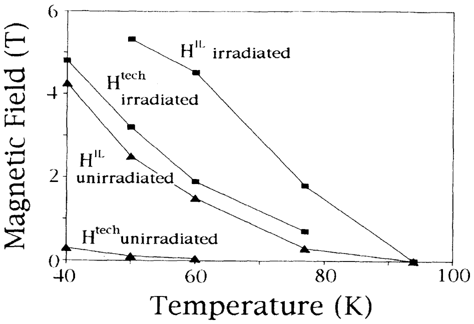


FIG. 4. Temperature dependence of the irreversibility magnetic field ($H^{IL}$) and the magnetic field for $J_c = 10^4$ A/cm$^2$ ($H^{tech}$) before (solid triangles) and after (solid squares) neutron irradiation. The preirradiation $H^{IL}$ matches very closely with that reported by Welp *et al.* (Ref. 3).

temperatures, including above 0.5 T at 77 K. Comparing Figs. 3(a) and 3(b), one finds that the $J_c$ enhancements from irradiation range from 1–2 orders of magnitude—a much larger enhancement than obtained in other oxide superconductors. As this indicates a lack of pinning centers before irradiation, we conclude that the grains and grain boundaries were initially clean. It is also interesting to note that the field dependence is also altered by irradiation, becoming concave down at low magnetic field and showing significantly less reduction in fields up to 1 T.

Figure 4 plots the irreversibility line, $H^{IL}(T)$, before and after irradiation. Additionally, the $H$-$T$ curves for fixed $J_c = 10^4$ A/cm$^2$ are shown ($H^{tech}$), indicating the cutoff line for superconducting applications. Note that at 77 K, irradiation increased $H^{IL}$ to 1.8 T, illustrating that, in the presence of pinning centers, $HgBa_2CuO_{4+\delta}$ is competitive with Y-Ba-Cu-O in magnetic behavior.

We have demonstrated that the recently discovered $HgBa_2CuO_{4+\delta}$ superconductor is capable of carrying large current densities when defects are introduced by neutron irradiation. As the magnetic properties are superior to Bi-Sr-Ca-Cu-O materials, and magnetic property measurements indicate that the grain boundaries may be clean, $HgBa_2CuO_{4+\delta}$ is potentially an important technological superconducting material.

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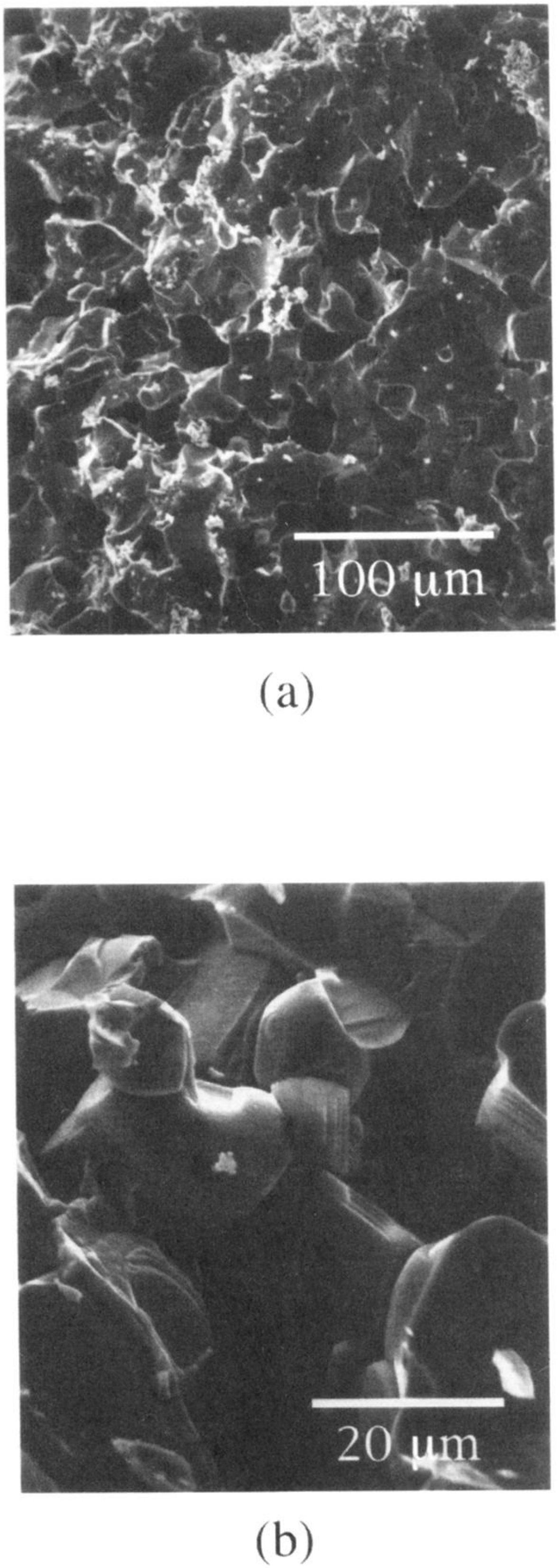

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