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Enhanced Supercurrents Above 100 K in Mercury Cuprates via Fission of Mercury

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Large-scale technological success of high-temperature superconductors will ultimately be decided by their capacity to sustain large critical current densities \( J_c \) in high magnetic fields. There are two principal factors controlling current conduction:\footnote{1,2} One is regions of weaker superconductivity (weak links) at the grain boundaries in polycrystalline materials.\footnote{1,2} Another is easy motion of magnetic vortices in the bulk - the result being energy dissipation and losses.\footnote{3} Each of these factors is a challenge to overcome, for their origin is intrinsic:\footnote{3} i.e. short superconducting coherence length \( \xi \), large anisotropy, large thermal fluctuations (related to high transition temperature \( T_c \)), and perhaps even a d-wave character of the superconducting ground state.\footnote{4} For these reasons, in spite of the highest \( T_c \)'s (\( >130 \) K), mercury cuprates \( \text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta} \) with \( n=1, 2 \) or 3 adjacent CuO layers (Hg-1201, -1212, or -1223) still have relatively low-lying irreversibility lines (supressed by a strong 2-D nature of the vortex structure), above which \( J_c \) vanishes. Here, we demonstrate a method by which we expand the useful range to \( T > 100 \) K (higher than in Y-, Bi-, or Tl-based materials\footnote{8-10}) and boost \( J_c \) by orders of magnitude in fields of several Tesla - namely fission of Hg nuclei within Hg-cuprates with energetic (0.8 GeV) protons. This technologically viable process allows 'doping' these cuprates with strongly pinning columnar defects\footnote{11}.

Pinning of magnetic vortices in high-\( T_c \) superconductors is demonstrably optimized with linear (columnar) defects.\footnote{12,13} One way to install an effective columnar defect structure is by irradiation with swift (\( \sim 1 \) GeV) heavy ions (e.g. Pb or Au\footnote{9,10,12}) - the result is a random array of almost parallel tracks of amorphized material, \( \sim 50-80 \) Å in diameter. Such columnar damage is known to shift the irreversibility line upward by tens of degrees and to expand the useful regime by several Tesla at high temperatures.\footnote{9,10,12} This method, however, is not useful for large-scale applications, since typical heavy ions penetrate only \( \sim 20-30 \) μm of the material.\footnote{14} Another way, which we have demonstrated recently,\footnote{11} is by irradiation with energetic (0.8 GeV) protons. Such protons can induce fission of \( ^{209}_{83} \text{Bi} \) nuclei in \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \) (Bi-1212).\footnote{11} This second method may be relevant for large scale applications, since protons penetrate large distances (half-meter) into the material.\footnote{14} Typical fission products in this case, \( ^{131}_{54} \text{Xe} \) or \( ^{86}_{36} \text{Kr} \) at \( \sim 100 \) MeV, are energetic enough to create extended, highly misaligned damage tracks.\footnote{15}
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Fission tracks in Bi-based cuprates significantly improve their current conduction in fields of a few Tesla, but the irreversibility line, above which the vortex matter "liquifies" and is very mobile, still hovers near liquid nitrogen temperature (77 K) for fields of 0.1 - 1 T and is ultimately limited by $T_c < 90$ K. So the question we asked is whether the fission process can be extended to other materials, in particular to Hg-cuprates - the highest $T_c$ materials to date. Experimentally, the Bi fission cross-section is about $\sigma \sim 250$ mbarns, a fraction (0.16) of the well known cross-section for $^{238}$U. Hg, on the other hand has not been fissioned experimentally. The calculated $\sigma$ values for $^{80}$Hg are within 80-110 mbarns - much smaller. We visualize the fission process as follows. Upon entering an Hg nucleus, a 0.8 GeV proton collides with nucleons and excites the nucleus. The nucleus distorts (like a droplet) and "boils off" nucleons (protons and neutrons) that carry off most of the forward momentum and jointly about 0.6 GeV of energy, culminating in a fission birth process. The remaining 200 MeV are split (statistically) between the fission daughters, which travel on opposite paths. Typical fission offspring in this case would be $^{90}$Zr and $^{41}$Nb at 95-100 MeV, with yet unknown threshold for columnar track formation. Another possible nuclear channel is spallation with a much larger $\sigma \sim 1000$ mbarns. This would evaporate several nucleons and convert $^{80}$Hg into a lighter particle with ~10-30 MeV recoil energy, e.g. $^{181}$Ta. The Ta recoil channel, however, is not expected to produce columnar tracks.

The polycrystalline Hg-cuprates used in this study were prepared following procedures described in Ref. 19 and 20. The Hg-1201 samples were prepared at ambient pressure, while the Hg-1212 and Hg-1223 were prepared using high (2-3 GPa) pressure synthesis method. Typical grain size was found by transmission electron microscopy (TEM) to be ~20\(\mu\)m. The onset of a sharp resistive transition was measured at $T_c \sim 95$K for Hg-1201, ~120K for Hg-1212, and ~127K for Hg-1223. The final materials were exposed to 0.8 GeV proton fluences of $1.3 - 1.7 \times 10^7$/cm$^2$ at Los Alamos National Laboratory using the proton beam from LAMPF at the WNR Facility. The proton fluence was determined to an accuracy of about 2% from proton induced $^{24}$Na activation of thin aluminum foils placed in front of each sample during its irradiation.

The resulting splayed columnar tracks in Hg-1212 recorded in TEM have track diameter in excess of 8 nm. The track length is estimated as follows. For a sample subjected to a calibrated fluence of $1.52 \times 10^{17}$/cm$^2$, the defect density from the TEM is $\approx 5.9 \times 10^{10}$/cm$^2$. Taking $\sigma_{Hg} = 110$ mbarns and Hg density $\rho_{Hg} \approx 5.3 \times 10^{21}$/cm$^3$ we get the density of fission events at $8.9 \times 10^{13}$ fiss./cm$^3$, which implies a mean track length of ~6.6\(\mu\)m for two fission fragments. This is consistent with the total range of 8 - 9.5\(\mu\)m for ~100 MeV Nb or Zr in Hg-1212 we obtain from TRIM Monte Carlo calculations. The electronic energy loss rate (~10 - 15 KeV/nm) threshold for the columnar track formation appears to be lower in Hg-cuprates than in Bi- or Y-based materials, plausibly due to their much higher resistance (a factor of 20-80) in the normal state.

The temperature dependence of the persistent current density $J$ in Hg-1212 before irradiation and with the fission density $8.9 \times 10^{13}$ fiss./cm$^3$ is shown in Fig. 1 for three different values of magnetic field. $J$ was estimated from hysteresis in magnetization via the critical state model using TEM determined grain size as the relevant current loop dimension. Before irradiation $J$ falls off very rapidly with temperature as the applied field is raised. At $\mu_0 H = 2$ T the fall off exceeds three orders of magnitude by the time...
\( \sim \frac{1}{2} T_c \) (~ 60 K) is reached, as is also seen in c-axis epitaxial Hg-1212 thin films. This precipitous fall is clearly arrested after irradiation and a spectacular expansion of the irreversible regime towards higher temperatures is evident in the Figure; at a 2 Tesla field and T \( \sim \)60 K, the increase in \( J \) is over two orders of magnitude. At low fields \( J \) remains measurable up to 110 K. Thus, the persistent current density becomes large in the regions above the irreversibility line of unirradiated samples.

Importantly, the mobility of vortices (dissipation) is reduced with splayed fission tracks not only in the irreversible regime, but also in the (liquid) flux-flow regime. This is apparent in Fig. 2, which shows (normalized) resistance vs temperature. The zero-field resistive transition (\( T_c \)) is left essentially intact by the fission process for this particular fluence, which corresponds to an equivalent matching field \( B_\Phi \sim 1.2 \) T. In finite fields, the two effects are apparent: (i) the onset of linear dissipation is shifted to higher temperatures and (ii) the flux-flow resistance (at any fixed T) is much lower after proton irradiation. The nonlinear dissipation (thermal creep) is reduced by a factor of \(~5\) near 40 K and more so near the irreversibility line \( H_{irr}(T) \) of the virgin material (inset of Fig. 2).

The expansion of the irreversible regime is illustrated in Fig. 3 which shows the irreversibility lines before and after irradiation. The irreversibility line was defined by (i) the onset of nonlinear current-voltage (I-V) characteristics with the criterion of 1\( \mu \)V/cm (main figure) and by (ii) the closing of the hysteresis loop \( M(H) \) at each temperature with the criterion of \( 5 \times 10^{-6} \) emu, corresponding to \(~200 \) A/cm\(^2\) (see inset of Fig. 3). The magnetic criterion underestimates the absolute \( T_{irr} \), but the relative shift is comparable for both methods. At 2 Tesla, for example, the upward shift of the irreversible regime is \(~30\) K - this moves \( H_{irr}(T) \) from below liquid nitrogen temperature to about 95 K. For fields \( H < 1 \) T, the irreversibility temperature \( T_{irr} \) shifts well above 100 K for \( B_\Phi \) of \(~1.2 \) T.

Such large effects are not limited to Hg-1212. This is illustrated in Fig. 4, which shows the relative shift of \( T_{irr} \) for Hg-1201, Hg-1212 and Hg-1223 with nominally the same field-equivalent defect densities. Indeed, we find a much larger relative shift \( \Delta T_{irr} \) (about 50 K at low fields) for a single-CuO-layer Hg-1201 and a comparable shift for a three-CuO-layer Hg-1223. The differences between the three Hg cuprates stem from the differences in their \( T_c \)'s that limit the absolute range (inset of Fig. 4), differences in material parameters, and anisotropies.

Thus, in Hg-based cuprates, both the enhancement of \( J \) at high fields and the shift of the irreversibility line with fission processing occurs at much higher temperatures than previously obtained for any other material. One possible reason for the effectiveness of the fission induced tracks in Hg-based superconductors at high temperatures is their relatively large effective diameter\(^3\). We surmise that this process is extendable to higher fluences, since for \( B_\Phi \sim 1.2 \) T, there is no measurable degradation in \( T_c \). So here we have the advantages of columnar tracks (with long portions of the vortex core confined by the defects\(^12\)) and with the pinning forces from individual defects adding directly, rather than statistically as with random defects) with a bonus of splay (dispersion)\(^24\) in the track directions. Splay can be beneficial in a highly anisotropic material\(^11\), it further boosts \( J_c \) by forcing entanglement of vortices and helps to limit
the escape of vortices from the tracks\textsuperscript{24}. This technique can be technologically relevant, since it is not limited by the short penetration range of heavier particles (or ions) and could permit modification of larger superconducting objects, such as magnets.

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**Figure Captions**

Fig. 1. The persistent current density $J$ vs temperature for Hg-1212 sample as synthesized and after processing with $1.52 \times 10^{17}/\text{cm}^2$ proton fluence, for three values of applied magnetic field. $J$ was obtained via Bean model from magnetization $M$ measured with a SQUID magnetometer\textsuperscript{11}. Bulk $J$ is enhanced to above $10^7/\text{A/cm}^2$. After irradiation $J$ becomes large above 100 K, well above the irreversibility line of unirradiated sample.

Fig. 2. Resistance $R$ (normalized to $R$ at $T_c$) before and after 0.8 GeV proton irradiation corresponding to the matching field $B_\Phi \sim 1.2$ T. While zero-field $T_c$ is unchanged, the flux-flow resistance is significantly reduced by this process, as shown for $\mu_0 H = 2$ T. Inset: The decay (creep) rate for currents is reduced with the fission induced columnar pins.

Fig. 3. Irreversibility lines of unirradiated and 0.8 GeV proton irradiated Hg-1212 (equivalent dose $B_\Phi \approx 1.2$ T) obtained from the onset of nonlinearities ($J_c$) in the I-V curves. The arrow points to a nearly 30 K shift at $\mu_0 H = 2$ Tesla upon irradiation. Inset: Irreversibility lines obtained from the collapse of magnetic hysteresis show a comparable shift (see text).

Fig. 4. Relative shifts of the irreversibility temperatures $\Delta T_{\text{irr}}(H)$ for Hg-1201, Hg-1212, and Hg-1223 with the same field-equivalent defect densities $B_\Phi$ of $\sim 1.2$ T. $\Delta T_{\text{irr}}$ as high as 50 K is observed. The shifts are relative to the virgin $T_{\text{irr}}(H)$, which are set by the $T_c$'s and by the anisotropies of the three Hg-cuprates (inset).
References


17. T.A. Gabriel, private communication.


Fig. 1 L. Krusin-Elbaum et al.
Fig. 3 L. Krusin-Elbaum et al.
Fig. 4 L. Krusin-Elbaum et al.