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# Magnetic irreversibility in $(Hg_{1-x}Re_x)Ba_2Ca_2Cu_3O_{8+\delta}$: effects of neutron irradiation

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**Abstract**

We studied the intragrain magnetic hysteresis of the superconductor $(Hg_{1-x}Re_x)Ba_2Ca_2Cu_3O_{8+\delta}$ ($x = 0.10$, 0.18 and 0.25) close to $T_c$. From the magnetization curves we determined the temperature dependence of the irreversibility field ($H_{irr}(T)$), and the field dependence of the magnetization curve width ($\Delta M(H)$). After irradiation to a fluence of $2 \times 10^{21}$ m$^{-2}$ ($E > 0.1$ MeV), the magnetization curves moderately increase their symmetry, and sizeable enhancements of $H_{irr}(T)$ and $\Delta M(H)$ are observed. Although the substitution $x = 0.18$ shows the best pinning properties before irradiation, $x = 0.10$ matches—or even surpasses—them afterwards. This suggests that, within our temperature and field window, neutron irradiation plays a far more important role than Re-substitution in enhancing the irreversible behavior in the Hg-1223 system. 

*Keywords:* HBCCO; Polycrystals; Hysteresis; Irreversibility line; Neutron irradiation

The superconductors of the Hg family have attracted growing interest due to their high critical temperature and low anisotropy, if compared with Bi- and Tl-based $HT_c$'s [1,2]. It has been shown that the partial substitution of Hg by Re increases the stability and pinning strength of the Hg-1223 phase, supposedly due to a decrease of the Cu–O interlayer distance, metallization of the interlayer planes, and added structural disorder [3–10]. On the other hand, neutron irradiation has proved to "symmetrize" the magnetization loops and improve pinning in "pure" Hg-1223 [11,12] and also in $(Hg_{0.85}Re_{0.15})Ba_2Ca_2Cu_3O_{8+\delta}$ [7].

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In this paper, a systematic study of magnetization loops at temperatures close to $T_c$ is presented for Re substitutions up to the observed solubility limit. The temperature dependence of the irreversibility field, and the field dependence of the magnetization curve width, indicate that the intermediate substitution is the best to enhance irreversibility in Hg-1223. We also present a study of neutron irradiation effects on our series of Re-substituted Hg-1223 polycrystals. Neutron irradiation improves pinning in a way not trivially correlated to the Re content: bulk pinning in $Re_{0.10}$ matches—or even surpasses—that of $Re_{0.18}$ after neutron irradiation.

Polycrystals of composition $(Hg_{1-x}Re_x)Ba_2Ca_2Cu_3O_{8+\delta}$ where obtained by stoichiometrically mixing $BaCO_3$, $CaCO_3$, CuO and $ReO_2$ high purity powders. The mixture was treated at 850 °C for 12 h under oxygen flow. Then, the material was crushed and pelletized under 1 GPa. The pellets were calcined twice at 930 °C for 15 h under oxygen flow, treated at 930 °C for 12 h in 20% oxygen and 80% argon flow. After HgO addition, the powder was pelletized uniaxially under 1 GPa in vacuum, and then introduced into a gold foil and both into a quartz tube with a filling factor of 1.1 g/cm$^3$ [6]. The tube was vacuum sealed, introduced inside an isostatic pressure furnace with 40 bar of argon, heated at 300 °C/h to 700 °C and at 120 °C/h to 850 °C, where it was treated for 15 h. Then it was cooled down at 120 °C/h to 30 °C. We synthesized the nominal substitutions $x = 0.10$, $x = 0.18$ and $x = 0.25$, which we will call "$Re_{0.10}$", "$Re_{0.18}$", and "$Re_{0.25}$" respectively. The first and third substitutions can be considered to be "extreme" ones, because $x = 0.10$ is relatively small and $x = 0.25$ is the solubility limit for Re. The $c$-axis parameter determined from XRD on analogous samples decreased in the order $Re_{0.25}$, $Re_{0.10}$ and $Re_{0.18}$ [10]. Two contiguous bars of approximate dimensions $10 \times 1 \times 1$ mm$^3$ were cut from each pellet, so one of them was irradiated with neutrons to a fluence of $2 \times 10^{21}$ m$^{-2}$ ($E > 0.1$ MeV) (TRIGA reactor at the Atominstitut der Österreichischen Universitäten, Vienna) [13]. The irradiated bars will be called "$Re_{0.10}I$", "$Re_{0.18}I$" and "$Re_{0.25}I$", respectively. The magnetic measurements were performed in a Quantum Design SQUID model MPMS-5S, in the DC mode, using a scan length of 1 cm to minimize magnetic field inhomogeneities, and 10 scans per point to diminish the noise. The field was applied along the longest dimension of the bars. The samples were zero field cooled, and the average speed of the field ramp was ≈1 mT/s in the case of magnetization loop measurements. The onset $T_c$ values for $Re_{0.10}$ ($Re_{0.10}I$), $Re_{0.18}$ ($Re_{0.18}I$) and $Re_{0.25}$ ($Re_{0.25}I$) (also measured in the SQUID) were 133.2 K (132.1 K), 133.1 K (132 K) and 132.6 K (131.7 K), respectively. Clearly, the neutron irradiation had little effect on $T_c$.

Fig. 1 displays the magnetization curves for samples $Re_{0.18}$ and $Re_{0.18}I$, which showed the best magnetic properties before irradiation. Considering the previous studies of the transport critical current density in these samples [14], we shall assume that all the features seen above a few mT, are intragranular. A cursory inspection of the figure

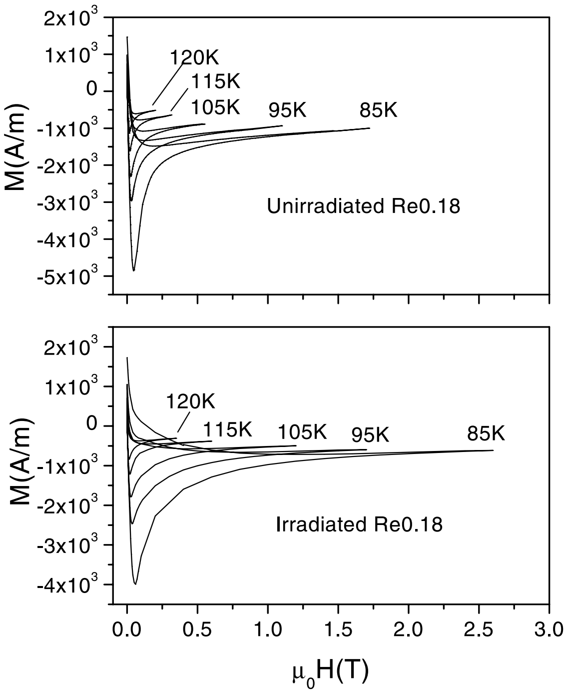


Fig. 1. Magnetization curves for the $Re_{0.18}$ sample before and after neutron irradiation. Error bars (almost totally associated to the uncertainty in the measurement of the sample dimensions) are not displayed.

indicates a strong asymmetry of the curves before irradiation as well as quite similar slopes for increasing and decreasing fields in the region above 1 T, which suggests a sizeable reversible component in the magnetization. After irradiation, the curves indicate an increase in the contribution of the irreversible magnetization probably associated with improved bulk pinning. This is not surprising if we take into account that the type of neutron irradiation we have applied, produces (at least in other $HT_c$ systems) collision cascades i.e., spherical defects of a few nanometers diameter, with a density of $10^{22}$ $m^{-3}$ at this fluence [15,16].

Fig. 2 presents the temperature dependence of the irreversibility line for our samples, defined as the point at which the increasing and decreasing branches of the magnetization loop split, with a criterion of 25 A/m. $Re_{0.18}$ displays the highest values of $H_{irr}$ for the unirradiated samples, as expected from XRD data (assuming the simple criterion that a small interlayer distance produces high irreversibility) [10]. The irradiation provokes an increase of $H_{irr}(T)$ for all the samples, particularly $Re_{0.10}$. The $H_{irr}$ versus $1 - T/T_c$ plot (based on the raw data shown in the inset) indicates that the temperature dependence of all our samples can be described by $H_{irr}(T) \sim (1 - T/T_c)^n$. It is generally accepted that $n = 2$ implies 3D vortex lines, while $n = 4$ has been associated with 2D vortices in the case of Hg-1223 (see, for example, Ref. [12]). In our case, $n$ moves in the range 1.5–1.9, which suggests that our vortex lines behave tri-dimensionally, regardless Re content or irradiation conditions. Quite similar values of $n$ have been observed both in the Hg-1212 and Hg-1223 systems, with a crossover to $n \sim 4$ for higher fields and lower temperatures, indicative of 2D vortices [17,12]. These $n$ values lie between the ones typically reported for YBCO and BSCCO materials, suggesting a correlation between vortex dimensionality and interlayer spacing. This gives additional evidence to the fact that, in our system, the smaller interlaying spacing and metallization associated to Re-doping [3–5] should enhance the 3D behavior of the vortex lines. After neutron irradiation, our exponents maintain approximately the same values, as expected after the addition of large pinning centers. It is worth mentioning that, in the case of non-doped Hg-1223 samples, neutron irradiation has demonstrated even to extend the 3D vortex region to lower temperatures and higher fields [12].

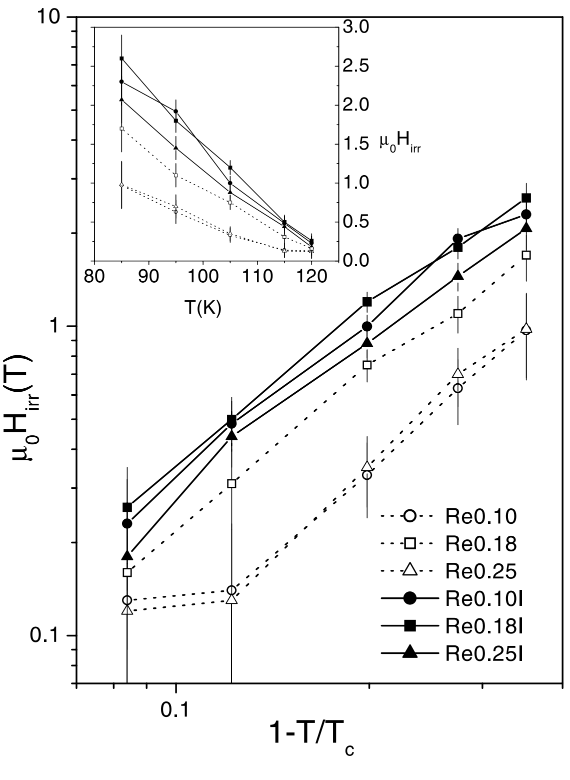


Fig. 2. Temperature dependence of the irreversibility field plotted in a way convenient to evaluate vortex dimensionality. The raw data is shown in the inset.

Fig. 3a shows the field dependence of the magnetization loop width versus the applied field taken at 105 K, i.e., approximately halfway our temperature range (similar plots at other points within our temperature window show qualitatively similar results). All the plotted values correspond to fields higher than the minimum of the magnetization curves. If we assume a bulk pinning scenario and apply the Bean's critical state model [18], this graph roughly represents the field dependence of the intragrain critical current density. While a moderate improvement is observed for $Re_{0.25}$ and $Re_{0.18}$, a stronger increase of the magnetization width takes place for $Re_{0.10}$ after irradiation.

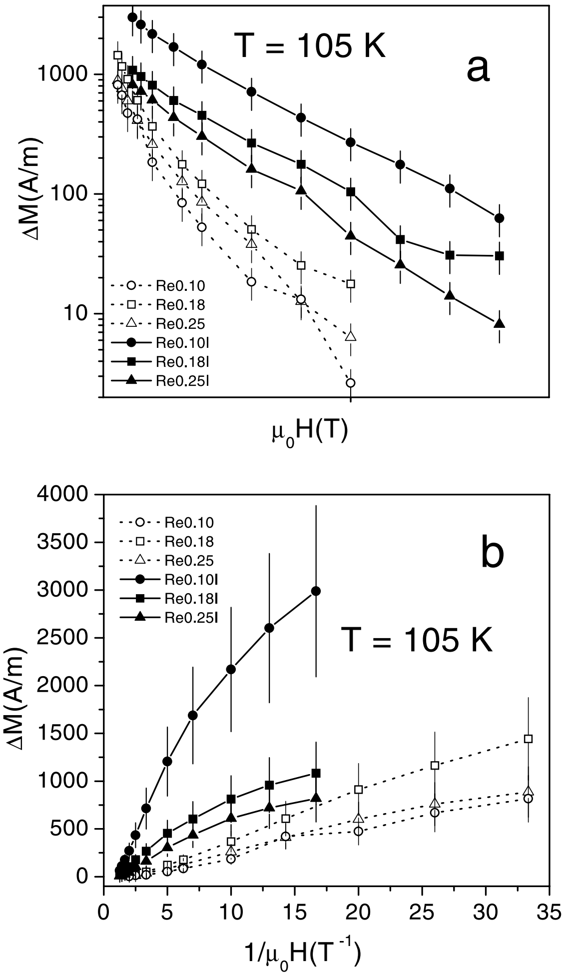


Fig. 3. (a) Field dependence of the width of the magnetization curves at $T = 105$ K. (b) Same data as (a), plotted against the inverse of the applied field.

Figs. 2 and 3a suggest that the action of neutron irradiation on the irreversible behavior of Re-doped Hg-1223 polycrystals is far more effective than the effects of Re doping. Since no data is available on the precise geometry and distribution of defects created by neutron irradiation in the Re-doped Hg-1223 system, only speculative arguments can be given to explain the stronger improvement of the irreversibility line observed in $Re_{0.10}$ and $Re_{0.25}$, if compared with $Re_{0.18}$. Considering that the pinning differences before irradiation cannot be related to the dimensionality of the vortex lines (they are 3D for all the dopings, as suggested above), and assuming that the interaction between neutrons and Re atoms is roughly similar to that for other elements found in high $T_c$ superconductors, the differences could be related to the defect structure of the samples prior to irradiation. If that is the case, we can assume that the defect density for $Re_{0.18}$ before irradiation (typically 2D defects on the *ab* planes such as stacking faults and dislocation loops bordered by partial dislocations [7]) was quite higher than those for $Re_{0.10}$ and $Re_{0.25}$, but much lower than that associated to neutron irradiation. We can then think that the pinning centers produced by neutron irradiation are so effective, that they "mask" the pre-irradiation pinning differences. The particularly strong effect on $Re_{0.10}$ might be related to some kind of interaction of the neutron-induced cascades with the "as-grown" pinning centers, which tend to improve in a lesser degree the samples with better initial pinning. The ample scope of such possible interactions is well illustrated in recent studies of the influence of neutron irradiation on the "fishtail" feature of 1–2–3 crystals [19].

Fig. 3b displays a $\Delta M(1/H)$ characteristic extracted from the data shown in Fig. 3a. The behavior of the type $M \sim 1/H$ observed in the unirradiated samples is commonly interpreted in the literature as a fingerprint of the existence of surface barriers (see, for example, [8]), by assuming the theoretical result $M \sim H_p^2/H$, where $H_p$ is the field value at which the vortices start to penetrate the surface barrier [20,21]. However, it should be stressed that the $H_p$ values obtained from this assumption do not match with those extracted from the virgin magnetization curves, either defining $H_p$ as the point where the curves departs from linearity, or as the field value corresponding to the curve minimum, both of them commonly used in the literature [22,8]. Even the relative values of $H_p$ corresponding to different samples and irradiations do not match. Then, our irreversibility behavior is totally or, at least, partially associated to bulk pinning. In the case of the irradiated samples, the importance of bulk pinning is even higher, as suggests the clear non-linear behavior displayed by the corresponding curves in Fig. 3b.

The study here presented indicates that neutron irradiation strongly influences pinning in (Hg, Re)-1223 polycrystals and, at least in the high temperature and low field region under study, is more important than the doping itself.

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## References

[1] S.N. Pulitin, E.V. Antipov, O. Chmaissem, M. Marezio, Nature 362 (1993) 226.
[2] A. Shilling, M. Cantoni, J.D. Guo, H.R. Ott, Nature 363 (1993) 56.
[3] J. Shimoyama, S. Hahakura, K. Kitazawa, Y. Yamafuji, K. Kishio, Physica C 224 (1994) 1.
[4] J. Shimoyama, S. Hahakura, R. Kobayashi, K. Kitazawa, K. Yamafuji, K. Kishio, Physica C 235–240 (1994) 2795.
[5] O. Chmaissem, J.D. Jorgensen, K. Yamaura, Z. Hiroi, M. Takano, J. Shimoyama, K. Kishio, Phys. Rev. B 53 (1996) 14647.
[6] A. Sin, A.G. Cuhna, A. Calleja, M.T.D. Orlando, F.G. Emmerich, E. Baggio-Saitovich, S. Pinol, J.M. Chimens, X. Obradors, Physica C 306 (1998) 34.
[7] J. Krelaus, M. Reder, J. Hoffmann, H.C. Freyhardt, Physica C 314 (1999) 81.
[8] L. Fábrega, A. Sin, S. Pinol, J. Fontcuberta, D. Eyidi, J. Rabier, Physica C 325 (1999) 34.
[9] A. Sin, L. Fábrega, M.T.D. Orlando, A.G. Cunha, S. Pinol, E. Baggio-Saitovich, X. Obradors, Physica C 328 (1999) 80.
[10] M.T.D. Orlando, A. Sin, L. Fábrega, F. Alsina, A.G. Cunha, N. Mestres, A. Calleja, S. Pinol, F.G. Emmerich, L.G. Martínez, M. Segarra, X. Obradors, E. Baggio-Saitovich, Physica C 328 (1999) 257.
[11] Y.R. Sun, J.R. Thompson, J. Schwartz, D.K. Christen, Y.C. Kim, M. Paranthaman, Phys. Rev. B 51 (1995) 581.
[12] A. Wisniewski, R. Puzniak, J. Karpinski, J. Hofer, R. Szymczak, M. Baran, F.M. Sauerzopf, R. Molinski, E.M. Kopnin, J.R. Thompson, Phys. Rev. B 61 (2000) 791.
[13] H.W. Weber, H. Böck, E. Unfried, L.R. Greenwood, J. Nucl. Mater. 137 (1986) 236.
[14] E. Altshuler, R. Cobas, A.J. Batista-Leyva, C. Noda, L.E. Flores, C. Martínez, M.T.D. Orlando, Phys. Rev. B 60 (1999) 3673.
[15] M.C. Frischherz, M.A. Kirk, J. Farmer, L.R. Greenwood, H.W. Weber, Physica C 232 (1994) 309.
[16] M. Aleska, P. Pongratz, O. Eibl, F.M. Sauerzopf, H.W. Weber, T.W. Li, P.H. Kes, Physica C 279 (1998) 171.
[17] Z.J. Huang, Y.Y. Xue, R.L. Meng, C.W. Chu, Phys. Rev. B 49 (1994) 4218.
[18] C.P. Bean, Phys. Rev. Lett. 8 (1962) 250.
[19] M. Werner, F.M. Sauerzopf, H.W. Weber, A. Wisniewski, Phys. Rev. B 61 (2000) 14795.
[20] J.R. Clem, in: K.D. Timmerhaus, W.J. O'Sullivan, E.F. Hammel (Eds.), Proceedings of the 13th Conference on Low Temperature Physics (LT13), vol. 13, Plenum, New York, 1974, p. 102.
[21] L. Burlachkov, Phys. Rev. B 47 (1993) 8056.
[22] M. Reissner, Physica C 290 (1997) 173.