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Effect of 5.3-GeV Pb-ion irradiation on irreversible magnetization in Y-Ba-Cu-O crystals

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We report a dramatic change in the irreversibility line of Y-Ba-Cu-O crystals after irradiation with Pb ions. Near the transition temperature, following irradiation, the irreversibility temperature increases and the curvature of the irreversibility line changes sign. These changes are accompanied by a strong enhancement of critical current density and a decrease in flux creep rate. Pb irradiation induces damage in the form of amorphous tracks which penetrate throughout the thickness of the sample. We maintain that these defects are most efficient in terms of flux trapping and are responsible for the observed changes in irreversible magnetic features in the irradiated sample.

Enhanced flux pinning in irradiated high-temperature superconductors (HTSC) has been the topic of numerous recent studies.[1] Various types of radiation have been used[2-8] including photons, electrons, protons, neutrons, and ions. In most of these studies attention is focused on increasing critical current density J_c. Dramatic enhancement of J_c has been reported for neutron- and proton-irradiated $YBa_2Cu_3O_7$ crystals,[2,3] demonstrating the efficiency of irradiation techniques in inducing flux-trapping defects in high-temperature superconductors. Surprisingly, only a few articles[3-6] deal explicitly with the closely related topic of the effect of irradiation on the irreversibility line.

The irreversibility line (IRL) in the field-temperature (H-T) phase diagram of HTSC marks the boundary between the regions of reversible and irreversible magnetic behavior. It was first determined by conventional measurements of zero-field-cooled (ZFC) versus field-cooled (FC) magnetization.[9] In these measurements the irreversible temperature $T_{\text{irr}}(H)$ is the temperature above which the ZFC and the FC branches of the magnetization coincide; hysteretic behavior is observed below T_{irr}. Numerous experiments have demonstrated that T_{irr} is proportional to H^q with $q<1$, typically $q=0.6$–0.7. Later, several other techniques were employed for the determination of the IRL and, in particular, an ac technique was proposed, based on the measurement of the temperature and field dependence of the third-harmonic signal V_3 in the ac response.[10] It has been demonstrated[10] that the onset of V_3 is related to the irreversibility temperature $T_{\text{irr}}(H)$. This technique yields qualitatively similar IRL ($q<1$) though the field prefactor is somewhat larger, namely that the IRL is "steeper," demonstrating a frequency dependence[11] to the IRL.

If the irreversible magnetic behavior is a consequence of flux pinning, one would expect that the irreversibility line would also be affected by the addition of irradiation-induced pinning centers. Civale *et al.*[3] have recently found that the irreversibility line of $YBa_2Cu_3O_7$ crystals is largely independent of the defect density introduced by proton irradiation, even at defect levels which enhance the irreversible magnetization by more than an order of magnitude. As stated by Civale *et al.*, this result is difficult to reconcile with most existing theories. The IRL was also measured by Gupta *et al.*[6] for a dense melt-processed $Bi_2Sr_22CaCu_1O_8$ before and after irradiation with 400-MeV oxygen ions. Contrary to the conclusion reached by Civale *et al.*, Gupta *et al.* find that the irreversibility line shifts slightly to higher reduced temperatures after irradiation, thus confirming the pinning-dependent nature of the irreversibility line.

In this paper we report a dramatic change in the irreversibility line of $YBa_2Cu_3O_7$ crystals after irradiation with Pb ions. The Pb irradiation not only shifts this line to higher reduced temperatures but also changes its shape; the characteristic upward curvature of the IRL obtained before the irradiation is changed to downward curvature after irradiation. In addition, we show that these changes in the IRL are accompanied by a strong enhancement of J_c and by a reduction in flux creep rate. These results indicate that flux-pinning centers introduced by Pb irradiation are fundamentally different from those produced by proton irradiation.

Two thin plates of $YBa_2Cu_3O_7$ crystals of dimensions $940\times860\times20$ and $900\times900\times20$ μm^3 were used in these experiments. Sample preparation is based on the procedure described in Ref. 12. We refer to the two samples as No. 1 and No. 2, respectively. The transition temperature for both crystals, determined by magnetic measurements, is $T_c=93.3$ K. Crystal No. 1 served as a reference. The other one was irradiated at Grand Accélérateur National d'Ions Lourds (GANIL, Caen, France), with a beam of 5.3-GeV Pb ions, at room temperature. Measurements on this sample were done before and after irradiation. The flux was limited to 10^8 ions/cm^{2}s in order to avoid any warming effect during irradiation. The total fluence was 7×10^9 ions/cm^2. Ions were incident parallel to the *c* axis (normal to the flat surface).

In 5.3-GeV Pb irradiation, the main part of the ion energy is transformed into electronic excitations and ionization. The electronic energy loss is approximately 35 keV/nm, and continuous amorphous tracks are expected along the ion paths.[13,14] No structural investigation was performed on the samples of interest here. Nevertheless, high-resolution electron microscopy measurements[14] on sintered $YBa_2Cu_3O_7$, irradiated by Pb ions in the same run, revealed the formation of continuous latent tracks with diameter of order 7 nm which extend throughout the whole thickness of the sample. From these results we may conclude that after irradiation, the crystal consists of normal amorphous cylinders embedded in a superconducting matrix. These cylinders, which penetrate throughout the whole crystal, are of diameter $\simeq 7$ nm and $\simeq 1000$ Å apart.

dc magnetization measurements were carried out on both crystals, with field parallel to the *c* axis, using three independent techniques: (i) A vibrating sample magnetometer (VSM) was used for measuring magnetization loops at low and intermediate temperatures. (ii) A superconducting-quantum-interference-device (SQUID) susceptometer was used for measuring temperature and time dependence of the magnetization. Conventional ZFC and FC magnetization curves have been used for determining the IRL. (iii) A recently developed technique,[15] based on the use of a miniature Hall probe, has been employed for measuring magnetization loops and flux creep in the vicinity of T_c. In addition, we employed an ac technique,[10] at a driving frequency of 20 kHz, for measuring the temperature and field dependence of the third-harmonic signal V_3 in the ac response. The onset of V_3 is related[10] to the irreversibility temperature $T_{\mathrm{irr}}(H)$.

Figure 1(a) exhibits results of magnetization curves at 44 K, recorded with the VSM, for the reference sample (1) and for sample 2 after irradiation. The figure shows a large enhancement of the hysteresis loop upon irradiation, indicating a significant increase in flux pinning and critical current density.[16] A similar enhancement is observed in all temperatures up to T_c. This is demonstrated in Fig. 1(b) which describes the "width" of the hysteresis loop at 90 K for sample 2, before and after irradiation. For the measurements at 90 K we have used the local Hall-probe technique.

For determination of the changes in flux creep rate and the pinning potential, magnetic relaxation measurements were performed on a SQUID susceptometer. Figure 2(a) compares the time dependence of the remanent magnetization at 10 K for the reference sample and for sample 2 after irradiation. In these measurements the sample was cooled to 10 K in a field of 1 kOe parallel to the *c* axis and magnetization data were recorded, starting approximately 300 s after turning off this field. The data of Fig. 2(a) show a pronounced reduction in flux creep rate after irradiation. For an order of magnitude estimation of the pinning potential *U* we use the "linear" approximation[17] $U \simeq kT/S$ where *S* is the normalized creep rate. From Fig. 2 we find $U_0 \simeq 0.08$ eV for the reference crystal and $U_0 > 1$ eV for the irradiated one. We note that a similar reduction in relaxation rate has been reported for a proton-irradiated Tl-Ca-Ba-Cu-O crystal.[7]

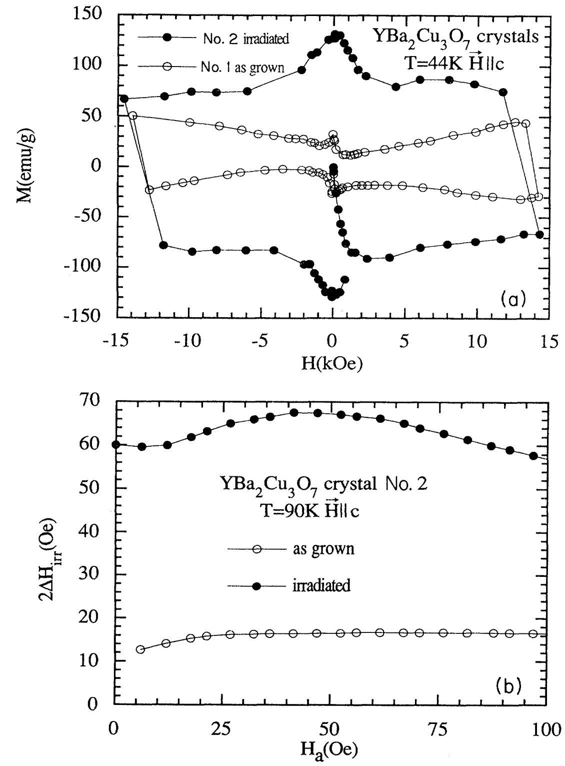

FIG. 1. Magnetization curves and width of hysteresis loop of $YBa_2Cu_3O_7$ samples before (open symbols) and after (solid symbols) irradiation. (a) Magnetization curves at 44 K, recorded with the VSM for samples 1 and 2. (b) Width of magnetization curves at 90 K for sample 2, measured by the local Hall-probe technique.

For relaxation measurements in the vicinity of T_c, we have used the local Hall-probe technique. Two samples, the reference one and the irradiated sample 2, were measured simultaneously. In these measurements we used the procedure: The sample is cooled in zero field to the measurement temperature (85 and 90 K in the present work). At this temperature the field is increased to its maximum value (400 Oe) and decreased back to a field *H* where the creep is recorded. Typical data for this procedure are shown in Fig. 2(b) which describe the magnetization as a function of time at 85 K for $H = 250$ Oe. The open symbols in the figures refer to the as-grown sample whereas the solid symbols refer to the irradiated sample. Before irradiation the normalized slope $S = d\ln(M)/d\ln(t)$ is $\simeq 0.023$. The irradiated crystal clearly exhibits much slower relaxation rates with normalized slope of approximately 0.013. In a later paragraph we derive the pinning energies from the values of *S*.

While the qualitative effect of Pb irradiation on the hysteresis and flux creep rate are similar for neutron and proton irradiation, its effect on the IRL is fundamentally different. The IRL before and after Pb irradiation was

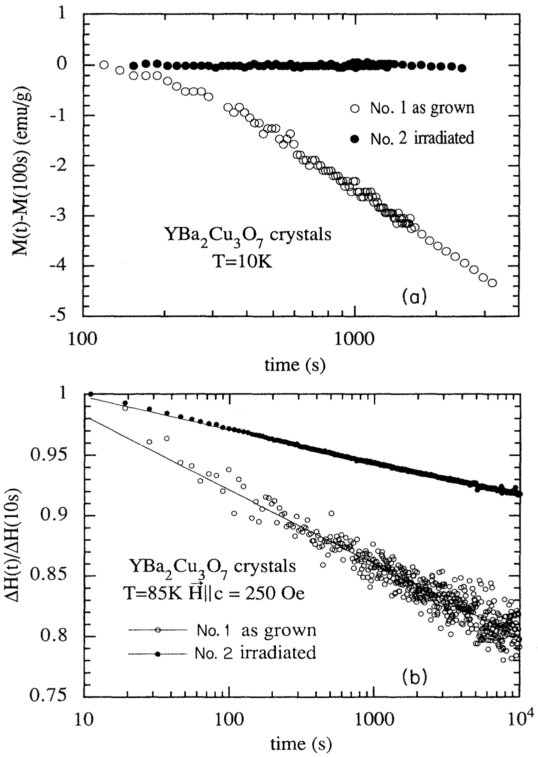

FIG. 2. (a) Time dependence of the remanent magnetization at 10 K for the reference sample (open symbols) and for sample 2 after irradiation (solid symbols). The $YBa_2Cu_3O_7$ samples were cooled to 10 K in a field of 1 kOe parallel to the *c* axis and magnetization data were recorded after turning off this field. (b) Magnetic relaxation of $YBa_2Cu_3O_7$ at 85 K, before (open symbols) and after (solid symbols) irradiation. The sample is cooled in zero field to 85 K, the field is increased up to 400 Oe and decreased back to 250 Oe where the creep is recorded.

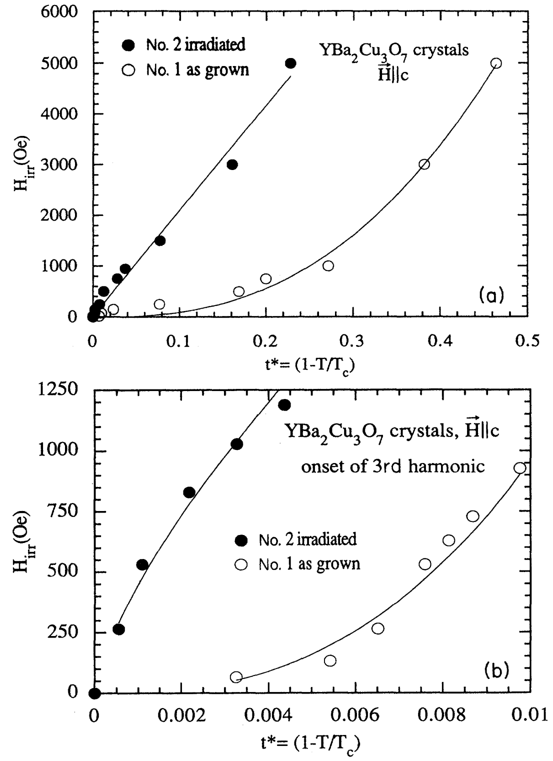

FIG. 3. The irreversibility lines (IRL) of $YBa_2Cu_3O_7$ samples before (open symbols) and after (solid symbols) irradiation. (a) The IRL determined by dc technique, from ZFC and FC magnetization curves. (b) The IRL determined from the onset of third harmonics in the ac response. The solid lines are theoretical fits, assuming a power-law dependence of the field on the reduced temperature.

determined by using both dc (SQUID) and ac techniques. In the dc technique $T_{\rm irr}$ is defined as the temperature above which the ZFC and the FC magnetization curves coincide within 0.5% of the measured signal. In the ac technique $T_{\rm irr}$ was determined from the onset of third harmonics in the ac response. The results of dc and ac measurements are summarized in Figs. 3(a) and 3(b), respectively. The solid lines in these figures are theoretical fits, assuming a power-law dependence of the field on the reduced temperature.[9] Both the dc and ac data indicate a dramatic change in the IRL following irradiation. We observe not only an upward shift of the IRL upon irradiation but also a drastic change in the power-law. Following irradiation, a change of the exponent from 0.6 to 1.3 and from 0.4 to 1.4 is indicated by the dc and ac data, respectively. The difference between the dc and ac data is probably due to the dependence of the IRL on the measuring frequency.

The overwhole impression obtained from the present experimental data is in favor of the conventional picture of bulk pinning due to defects or imperfections. The damage induced by Pb irradiation is well defined and, as stated above, may be described as continuous cylinders of diameter of $\simeq 7$ nm which penetrate throughout the whole thickness of the sample. These continuous, and linear, tracks provide frequent core pinning sites for each flux line along the total length of the line for $H \parallel c$. The pinning efficiency of such linear defects is well understood in light of the model discussed by Hylton and Beasley.[18] In their model the pinning energy of a vortex line of length l depends on l_p/l (l_p is the portion of the pinned length), and on the "tension" energy of distorted flux line. (A vortex line will decrease its free energy when it picks up a pinning site, but it will increase its free energy by "wandering" through the crystal in order to pick up pinning sites.[18]) The most favorable situation is expected for $l_p/l \simeq 1$ and for the minimum tension, i.e., a vortex which is pinned along its length, in the *c* direction. Pb irradiation is apparently capable of producing pinning centers which may be of maximum efficiency in terms of pinning energy.

As pointed out by Malozemoff and Fisher[19] normalized-magnetic-relaxation rates (S) exhibit a "universal" value of approximately 0.03 at moderate and high temperatures. This universality reflects the fact that relaxation rate is determined by the attempt frequency $1/t_0$ and not by the pinning energy. This prediction is made in the framework of collective flux pinning[20] or vortex glass[21] pictures, in which the logarithmic relaxation persists for observation times $t_{\rm obs}$ far above some critical time $t_{\rm cr}$

defined by the condition $\ln(t_{\rm cr}/t_0)=U/kT$. From Refs. 20 and 21 one may derive an approximate expression for the magnetic decay rate: $S=[U/kT+\mu\ln(t_{\rm obs}/t_0)]^{-1}$. In our experiment we observe a pronounced depression of S, even in the vicinity of T_c (85 and 90 K). In the low-temperature range we even observe a much more dramatic reduction in S. These results are consistent with a picture in which U is increased to such a level that $t_{\rm cr}$ is in the range of the observation time ($t_{\rm obs}$) even near T_c. By using the approximate expression for S we find $U \simeq 0.15$ eV at 85 K and $U>1$ eV at 10 K.

The shift of the IRL after irradiation is clear evidence that flux pinning is the origin of the IRL. This shift occurs after introducing new pinning centers of maximal efficiency, i.e., with U much larger than any of the preexisting defects in the unirradiated sample. It is not clear, however, what the origin is of the *qualitative* change in the *slope* of IRL after Pb ion irradiation. We propose that the new defects, which presumably are not produced by other irradiation techniques, are responsible for the observed changes in the IRL. In order to explain the apparent contradiction between our results and those of Ref. 3 we argue that J_c is determined by the density of pinning centers, whereas the IRL is determined by the temperature dependence of the largest U's. Pb irradiation induces dense defects with relatively large U values, thus causing changes in J_c as well as in the IRL. This is achieved by producing unique pinning centers which enable trapping of "linear" fluxons.

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