Magneto-resistance analysis of nanometer Al$_2$O$_3$ added Bi-2223 polycrystalline superconductors.

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Abstract. We report magneto-resistivity analyses on (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ (denoted as (Bi,Pb)-2223) granular samples added with nano-sized Al$_2$O$_3$ particles. Al$_2$O$_3$ is added to the precursor powders during the final sintering cycle of a multi-step preparation process. Resistivity measurements of 0.0 and 0.2 wt. % Al$_2$O$_3$ added (Bi,Pb)-2223 samples in applied magnetic field ranging from 0 to 250 mT have been compared and analyzed by the thermally assisted flux creep model. The effective activation energy in the Al$_2$O$_3$ added sample is higher than in free one. The vortex phase diagrams in the H-T space show that characteristic temperatures $T_g$ and $T_p$ shift to higher values with 0.2 wt.% Al$_2$O$_3$ addition.

1. Introduction
In our previous work [1], the results of the effect of nano-size Al$_2$O$_3$ (40 nm) addition on the microstructure and vortex pinning properties of polycrystalline (Bi,Pb)$_2$Sr$_2$Ca$_2$Cu$_3$O$_x$ have been presented. We have reported that the addition of a small amount (0.2 wt.%) of Al$_2$O$_3$ during the final processing of (Bi,Pb)-2223 superconductors, did not affect its formation but increased the critical current density $J_c$ and improved the $J_c$ (H) behavior in applied magnetic field. Transmission electron microscopy (TEM) and energy dispersive X-ray spectroscopy (EDS) analyses have shown the presence of nanometric Al–rich phase intergrowth within the superconducting matrix. The pinning force analysis proves that the enhancement of the vortices flux pinning in 0.2 wt. % Al$_2$O$_3$ added samples is mainly originated from the surface normal-like pinning centers.

The broad resistivity transition of high-Tc superconductors in a magnetic field has been a subject of great interest. This effect has been widely considered to be the result of the thermally activated flux motion of vortices and could be used to provide rich information about the properties of high-Tc superconductors at lower temperatures regime.

In the present paper, we discuss the physical mechanisms responsible of the broadening of the electrical resistivity $\rho$ (T,H) broadening of the Al$_2$O$_3$ nano-sized particles added samples under applied magnetic field. Then we investigate the broadening of the electrical resistivity transition with the thermally assisted flux creep model proposed by Anderson and Kim [2,3].

2. Experimental
Polycrystalline samples of (Bi,Pb)-2223 were elaborated by the solid-state synthesis route through a two-cycle annealing process. Details of the sample preparation and characterization have been described previously in Ref. [1], and herein we give a brief description. Al$_2$O$_3$ particles with 40 nm in diameter were added during the second thermal cycle to the precursor powder by mixing and hand
grinding powders. Then, powder mixtures were pressed into pellets under a uniaxial pressure of 1GPa. The pellets were carefully cut into bars shaped samples with active cross sections for current flow of 0.3 mm². Specimens were sintered at 830°C for 72 h in air. The structure and phase purity of the powder sample ground from sintered pellets were examined by powder XRD. We have found that both samples are mainly composed of (Bi,Pb)-2223 and have similar compositions, consisting of a mixture of the (Bi,Pb)-2223 phase (~90%) with extra phases, such as Bi-2212(~9%) and a small quantity Ca₂PbO₄ (~1%). The temperature dependence of the electrical resistivity for different values of applied magnetic field, ρ(T, H), of samples, was measured by using the standard dc four-probe technique with a fixed applied transport current of 40µA. The applied magnetic field in the range 0 to 250 mT was applied perpendicular to the thickness and to the excitation current that was injected along the major length of samples. The acquisition of the data close to the transition was obtained with the temperature varying in rates of 0.5 K/min.

3. Results and discussions
To further investigate vortices flux pinning enhancement, samples temperature dependence of the in-plane resistivity out of and under various magnetic fields perpendicular to their surface are measured. The ρ(T, H) transition curves are characterized by two distinctive sections; the upper section (T > Tc) characterizes the intra-granular transition and the lower one (T < Tc) characterizes the grain-boundaries network or inter-granular effects, which are considered to be weak Josephson type links [4]. Tc is the peak temperature of the ρ(T) curve (Inset Fig.1). In order to compare the experimental data ρ(T, H) with theoretical predictions in the section T < Tc we have used the thermally-activated flux-creep model proposed first by Anderson [2] and Anderson and Kim [3]. The broadened resistivity curves for high-Tc superconductors can be expressed as:

\[ \rho(T, H) = \rho_p \exp\left(-\frac{U_{\text{eff}}(T, H)}{k_B T}\right) \]  \hspace{1cm} (1)

and

\[ U_{\text{eff}}(T, H) = U(H)U(T), \]

where \( k_B \) is the Boltzman constant, \( \rho_p \) is the pre-exponential factor independent of the applied magnetic field and \( U_{\text{eff}}(H, T) \) the effective activation energy. The magnetic field \( U(H) \) dependence can be expressed as [5]:

\[ U(H) = \left(\frac{T_c(0)}{T_g(H)} - 1\right)^{-1}, \]  \hspace{1cm} (2)

where \( T_c(0) \) is the zero field critical temperature and \( T_g(H) \) is the glass transition temperature. i.e \( T_g(H) \) can be extracted by linear extrapolation of the plot of \( (d \ln(\rho/\rho_p))/dT)^{-1} \) versus T (Fig.1).

**Figure1.** \( (d \ln(\rho/\rho_p))/dT)^{-1} \) versus T at magnetic field \( \mu_0H = 250 \) mT. \( T_g \) is shown by arrow. Inset: dp/dT versus T.
Considering Eq 2 and Eq (1), we find that the temperature dependent \( U(T) \) is given by:

\[
U(T) = - \left[ k_B T \left( T_c(0) - T_g(H) \right) / T_g(H) \right] \ln(\rho / \rho_p)
\]  \( \text{(3)} \)

Previous studies suggested that the temperature dependent \( U(T) \) form is \( U(T) = U(0)(1 - T/T_c)^n \), with the parameter \( m \) usually chosen to be 3/2 [6], 2 [7] or 1 [8] for the high-Tc superconductor materials. It can be noted that the double transition anomaly of \( \rho(T, H) \) observed generally at \( T < T_c \) in polycrystalline materials is attributed to the extrinsic effect. So this feature should be properly taken into account when analyzing dissipation phenomena and the \( T_c \) must be a fitting parameter noted \( T_p \).

One can extract therefore the magnetic field dependence of the activation energies \( U(H) \) from the slope of \( \ln(\rho/\rho_p) \) vs \((1 - T/T_p)^m / T\) curves. It should be noted that for each sample the resistivity \( \rho_p \) at temperature \( T_p \) is constant for all the applied magnetic fields. \( m \) and \( T_p \) parameters were determined from fitting experimental curves \( U(T) \) (Eq (3)). Typical result of \( U(T) \) for free and 0.2 wt.% \( \text{Al}_2\text{O}_3 \) added samples is presented in Fig.2. \( U(T) \) curve of 0.2 wt.% \( \text{Al}_2\text{O}_3 \) added sample is displaced towards the high temperature.

Moreover, the fitting parameter \( m \) is found to be 1.5 ± 0.15 (Inset Fig.2) in all samples, which corresponds to the vortices lattice 3D behaviors [9]. The observed deviation of the experimental and calculated curves at temperature noted \( T_{BP} (T_{BP} < T_p) \) may be originated from a different dissipative process. This crossover temperature \( T_{BP} \) was observed by Bahilla et al. [10] in the granular \( \text{Bi}_{1.7}\text{Pb}_{0.3}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x \) superconductors. Above \( T_{BP} \) the order parameter fluctuation seems to dominate the cause of dissipation phenomena and below \( T_{BP} \) the vortex dynamics seems to dominate the cause of dissipation. In the case of \( \text{Al}_2\text{O}_3 \) added sample \( T_{BP} \) values are much higher than for the free one, in all range of applied magnetic field.

Since we are interested by the dynamics of vortex, we limited our discussion in the temperature range \( T_g < T < T_{BP} \).
Fig. 3 shows the variation of $\ln(\rho/\rho_0)$ vs $(1-T/T_p)^m/T$ at various magnetic fields for free and 0.2wt. % Al$_2$O$_3$ added samples. One can observe at a fixed magnetic field a linear behavior below the $T_B$ temperature as indicated by a solid line.

![Figure 3. Plots of the low temperature $\ln(\rho/\rho_0)$ versus $(1-T/T_p)^m/T$ at various magnetic fields (a) free and (b) 0.2wt. % Al$_2$O$_3$ added samples.](image)

$U(H)$ can be obtained from the slope of plots of Fig. 3 for $m = 3/2$. The activation energy $U(H)$ of 0.2wt.% Al$_2$O$_3$ added sample are much higher than for the free one, in all range of applied magnetic field, which confirm the strong pinning effect of vortices in this sample (see Fig.4).

![Figure 4. Magnetic field dependence of the activation energy $U(H)$ for free and 0.2wt. % Al$_2$O$_3$ added samples.](image)

According to the above results, the field dependence of the characteristic temperature $T_s$ and $T_p$ of free and 0.2 wt.% added Al$_2$O$_3$ samples are summarized in Fig. 5, then we obtain the vortices phase
diagram in the H-T space. From this phase diagram, one can see that $T_g$ and $T_p$ shift to higher temperature values for 0.2 wt.% Al$_2$O$_3$ added sample. The obtained results confirm the role of nano-sized Al-rich phase imbedded in the superconducting matrix in the improvement of the phenomenon of vortex pinning properties observed by measurements of the density of electric current under magnetic field.

**Figure 5.** Vortex phase diagram for free and 0.2 wt. % Al$_2$O$_3$ added samples.
Open symbol: temperature $T_g$
Solid symbol: temperature $T_p$

### 4. Conclusion

We have studied the magneto-resistivity properties of Al$_2$O$_3$ added (Bi.Pb)-2223 polycrystalline samples in applied magnetic field. The analysis of resistive transitions has shown that the dissipation phenomenon follows the thermally activated flux creep model in $T_g < T < T_B$ region. The vortex phase diagram in the H-T spaces show that $T_g$ and $T_p$ shift to higher temperature values with 0.2 wt.% Al$_2$O$_3$ addition. The obtained results confirm the role of nano-sized Al-rich phase imbedded in the superconductor matrix in the improvement of the phenomenon of vortex pinning properties observed by measurements of density of current under magnetic field.

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