Weak field magnetic susceptibility fluctuations above the superconducting transition of La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ (RE=Y, Sm, Gd, Dy, Ho, Yb) superconductor

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We report detailed experimental results of magnetic susceptibility for the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ superconducting system, above and close the superconducting transition. The data were obtained from 5 K up to 300 K, and applied field of 0.1 T, correspond to the so-called weak magnetic field limit. In this limit, the excess of magnetization is associated with the fluctuations of the vortex lines positions. These effects of thermal fluctuations on $M_{ab}$ can be quantified through the known “excess of diamagnetism” $\Delta \chi_{ab}$, but $T_c/T_C$. In this work we present a study of magnetic susceptibility fluctuations in the limit of weak magnetic fields, above the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ ($\text{RE}=\text{Y}, \text{Sm}, \text{Gd}, \text{Dy}, \text{Ho}, \text{Yb}$) high superconducting material. Samples were synthesized by the standard solid state reaction. The best adjustment in the experimental data, in the limit of weak magnetic field, of $\Delta \chi/T$ in function of the reduced temperature allowed to obtain the values of $A_S$ (diamagnetism of Schmidt) and $B_{LD}$ (parameter LD), in each one of the samples of the systems La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$. For the fluctuation analysis, we use the concept of excess of magnetization, based on the Lawrence-Doniach model (LD), which allowed to calculate the diamagnetism induced by thermal fluctuations in the normal state, in the vicinities of critical temperature $T_{c0}$ (74, 58, 74, 55, 60, 46 K, for RE=Y, Sm, Gd, Dy, Ho, Yb respectively). It was demonstrated, by means of the analysis of the thermal fluctuations, that the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ system shows an excellent 2D scaling behavior ($B_{LD}$).

Keywords: Cuprate superconductors; fluctuations; thermodynamic properties.

Reportamos resultados experimentales de medidas de susceptibilidad magnética para el sistema superconductor La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$, arriba y abajo de la transición superconductora. Los datos fueron obtenidos desde 5 K hasta 300 K, y un campo aplicado de 0.1 T, correspondiente al llamado límite de campo magnético débil. En este límite, el exceso de la magnetización es asociado con las fluctuaciones de la posición de las líneas de vórtice. Estos efectos de las fluctuaciones térmicas en $M_{ab}$ pueden ser cualificados a través de conocido “exceso de diamagnetismo” $\Delta \chi_{ab}$, en $T_c/T_C$. En este trabajo presentamos un estudio de las fluctuaciones de la susceptibilidad magnética en el límite de campo magnético débil, para el material superconductor La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ ($\text{RE}=\text{Y}, \text{Sm}, \text{Gd}, \text{Dy}, \text{Ho}, \text{Yb}$). Las muestras fueron sintetizadas por el método estándar de reacción de estado sólido. El mejor ajuste en los datos experimentales, en el límite de campo magnético débil, de $\Delta \chi/T$ en función de la temperatura reducida permitió obtener los valores de $A_S$ (diamagnetismo de Schmidt) y $B_{LD}$ (parámetro LD), en cada una de las muestras del sistemas La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$. Para el análisis de las fluctuaciones, usamos el concepto de exceso de magnetización, basados en el modelo de Lawrence-Doniach (LD), lo cual permitió determinar el diamagnetismo inducido por las fluctuaciones térmicas en el estado normal, en las vecindades de la temperatura crítica $T_{c0}$ (74, 58, 74, 55, 60, 46 K, para RE=Y, Sm, Gd, Dy, Ho, Yb respectivamente). Esto fue demostrado, por medio del análisis de las fluctuaciones, que muestra un excelente comportamiento scaling 2D ($B_{LD}$) para el sistema La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$.

Descriptores: Cupratos superconductores; fluctuaciones; propiedades termodinámicas.

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1. Introduction

High-temperature copper-oxide superconductors (HTSC) present important thermal fluctuation effects on both sides of superconducting transition. One of best adapted observables to the thermal fluctuations study in HTSC is magnetization, $M_{ab}(T,H)$, for magnetic fields, $H$, applied perpendicular to the CuO$_2$ planes, which is the so-called weak-amplitude limit [1-2]. High precision data of the influence of the superconducting order parameter fluctuations on the magnetization, $M(T)$, in the weak magnetic field limit, will contribute to the understanding of the superconducting transition in copper-oxide materials [2].

In the mean field-like regime, above superconducting transition, the fluctuation-induced magnetization, $\Delta M$, it is used to check some general aspects of the superconducting pairing state and to obtain useful relationships between some of the characteristics lengths arising in the Ginzburg-Landau descriptions of the superconducting transition [3]. Thermal fluctuations effects on $M_{ab}(T,H)$ may be quantified through the so-called magnetization excess, $\Delta M_{ab}(T,H)$, defined as

$$\Delta M_{ab}(T,H) \equiv M_{ab}(T,H) - M_{abN}(T,H) \quad (1)$$

where $\Delta M_{abN}(T,H)$ is the magnetization associated with the normal contributions, i.e., the sample magnetization if the su-
perconducting transitions were absent, and which may be approximated by extrapolating through the transition magnetization measured well above \(T_{C0}\), in a temperature region where the thermal fluctuation effects become negligible [3].

Above \(T_{C0}\), \(\Delta M_{ab}(T,H)\) is only due to thermal fluctuations, which create non equilibrium cooper pairs with a finite lifetime. In turn, these fluctuations induced Cooper pairs lead to appearance of shielding currents close the transition, which round down \(M(T)\) [4].

The aim of this work is to analyze susceptibility fluctuation effects in the weak-amplitude limit of high temperature superconducting oxide La\(_{0.5}\)RE\(_{0.5}\)Ba\(_2\)Ca\(_2\)Cu\(_3\)O\(_{7-\delta}\) (RE=Y, Sm, Gd, Dy, Ho, Yb), by using the Lawrence-Doniach (LD) two-dimensional (2D) model [5]. Our results reveal that fluctuations in this material have 2D character and respective critical parameters are determined from the excellent adjusting of experimental data with the LD model.

2. Experimental

Samples of La\(_{0.5}\)RE\(_{0.5}\)Ba\(_2\)Ca\(_2\)Cu\(_3\)O\(_{7-\delta}\) (RE=Y, Sm, Gd, Dy, Ho, Yb) material were synthesized by the standard solid state reaction recipe, from Aldrich precursor powders of CaO (99.99%), BaO (99.99%), CuO (99.99%) and RE\(_2\)O\(_3\) (99.9%). Powders were mixed, ground, palletized and calcined at temperature of 900 °C for 24 h. Then, material was reground, pressed as circular discs and sintered at 900 °C for 36 h, with two intermediate pulverizations.

Crystalline structure was studied by x-ray diffraction experiments, with a nickel-filtered Cu-K\(_\alpha\) radiation (\(\lambda=1,5406\) Å) of a SIEMENS D5000 equipment. Rietveld refinement by using GSAS code [7] was performed to determine the effective position of Ba and Ca ions into the structure. Analysis shows that material crystallizes in a tetragonal perovskite structure, space group P4/mmm, with lattice parameters \(a=3.859(4)\) Å and \(c=11.645(8)\) Å (Fig. 1). Magnetization measurements, \(M(T)\) and \(M(H)\), were performed through a 2000-model Quantum Design Magnetic Properties Measurement System (MPMS).

3. Results and discussion

For measurements with H perpendicular to ab planes and in the weak magnetic field limit, above \(T_{C0}\), both measured and the background magnetization arising in \(\Delta M_{ab}\) of Eq. (1) depend linearly of the magnetic field amplitude. Therefore, in this case, to characterize the thermal fluctuation effects on \(\Delta M_{abN}(T,H)\) it will be useful instead of \(\Delta M_{ab}\) the in-plane diamagnetism excess (also called in-plane fluctuation induced diamagnetism), \(\Delta \chi_{ab}(\varepsilon)\), defined as \(\Delta \chi_{ab}(\varepsilon) = \chi_{ab}(\varepsilon) - \Delta \chi_{abN}(\varepsilon)\), where \(\chi_{ab}(\varepsilon)\) are the measured and the background susceptibilities in the weak magnetic field limit, respectively, and \(\varepsilon=(T-T_{C0})/T_{C0}\) represents the reduced temperature. In planar superconductors with two superconducting layers per periodic length and with two different Josephson coupling strengths, \(\gamma_1\) and \(\gamma_2\), between adjacent layers, in the mean-field-like temperature region and in the weak magnetic field limit \(\Delta \chi_{ab}(\varepsilon)\) is given by

\[
\frac{\Delta \chi_{ab}(\varepsilon)}{\varepsilon} = -N_e(\varepsilon) \frac{A_s}{\varepsilon} \left(1 + \frac{4B_{LD}}{\varepsilon}\right)^{-1/2}
\] (2)

where \(N_e(\varepsilon)\) is an effective number of independent fluctuating planes per periodicity length, \(A_s \equiv \pi \mu_0 k_B \xi_{ab}(0)^2/\Theta_0^2\) is the Schmidt diamagnetism amplitude and \(B_{LD} = [\xi_{ab}(0)/\xi_{ab}^2(0)]^2\) is the Lawrence-Doniach parameter governing the dimensionality of thermal fluctuations of the order parameter [4]. Due the structural characteristics of La\(_{0.5}\)RE\(_{0.5}\)Ba\(_2\)Ca\(_2\)Cu\(_3\)O\(_{7-\delta}\) materials, we consider that this model is applicable for the study of diamagnetic fluctuations in them. To further analyze our experimental results on the grounds of Eq. (2), we must to use explicit expressions and values for \(N_e(\varepsilon)\) and \(g\). The approximation supposes that the strength of the Josephson coupling of each plane with its neighbor plane is the same for all planes, independently of their location. In that case, \(N_e(\varepsilon)\) takes the form [3]:

\[
N_e(\varepsilon) = 2 \left(1 + \frac{B_{LD}/\varepsilon}{1 + 4B_{LD}/\varepsilon}\right)^{1/2}
\] (3)

In the 2D limit each plane fluctuates independently \((\xi_\varepsilon(\varepsilon) << s)\) and \(B_{LD} << \varepsilon\). In the 3D limit \((\xi_\varepsilon(\varepsilon) >> s)\) and \(B_{LD} >> \varepsilon\). To compare these results with our experimental data, it is useful to rewrite the Eq. (2) by using Eq. (3) to obtain:

\[
\frac{\Delta \chi_{ab}(\varepsilon)}{\varepsilon} = -2A_s \left(1 + \frac{4B_{LD}}{\varepsilon}\right)^{-1/2}
\] (4)
Magnetic susceptibility ($\chi = M/H$) of the 
\[ \text{La}_{0.5}\text{Dy}_{0.5}\text{BaCaCu}_3\text{O}_{7-\delta} \]

sample measured with an applied magnetic field of 0.1 T is shown in Fig. 2. The solid line represents the contribution of the magnetic susceptibility in the normal region.

For $\Delta \chi_{abN}(T,H)$ measurements, samples were cooled down up to 5 K in absence of magnetic field by the known zero field cooling (ZFC) recipe. Then, magnetic field was applied and data were taken as the temperature was increased up to 250 K. The applied magnetic field of $H=0.1$ T corresponds to the weak magnetic field limit, where each order parameter component is expected to fluctuate independently. This limit can be defined by the condition [4]:

$$l_H \equiv \left( \frac{\hbar}{2e\mu_0 H} \right)^{1/2} \gg \xi_{ab}(\varepsilon) \tag{5}$$

The value of $T_{C0}$, which was used to numerically calculate the reduced temperature, was obtained by the extrapolation of the linear region of $T/\Delta \chi$ as a function of the La$_{0.5}$Dy$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ sample. This procedure has been used by other authors for two-dimensional systems as Bi:2232 and Bi:2212 [2]. The diamagnetism excess above $T_{C0}$ is presented in Fig. 3, for La$_{0.5}$Ho$_{0.5}$BaCaCu$_3$O$_{7.5}$ sample. The best adjustment in the experimental data of the Eq. (2) it allowed to determine the values of $B_{LD}$ and $A_S$ parameters [4].

The obtained values for the $B_{LD}$ and $A_S$ parameters, as well as the values of $T_{C0}$ and $T_C$ are detailed in Table I. From Eq. (3) and the value of separation of the plane $s \approx 11.7$ Å [7], we obtain $\xi_C(0)$ for all samples, which represents the coherence length associated to the fluctuational regime along c crystallographic direction. This value is approximately similar to experimentally obtained parameters of Bi:2212 and Bi:2232 superconducting systems. In the same form, by using Eq. (4), we calculate the correlation length of fluctuations in the ab planes, $\xi_{ab}(0)$. These results constituted the first evidence of the two-dimensional anisotropic character of the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ (RE=Y, Sm, Gd, Dy, Ho, Yb).

Inset of Fig. 4 represents a linear regime in a plot of $\log(\Delta \chi/T)$ as a function of $\log(\varepsilon)$. Slope of the linear behavior in the inset of figure 4 is $x=-1$. In order to establish the two-dimensional anisotropic character of the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ superconducting material, we consider the 2D limit. Then we use the previously proposed expression $B_{LD} < \varepsilon$ or equivalently $\xi_c(\varepsilon) << s$. Therefore, we can write Eq. (2) as [7,8]:

$$\frac{\Delta \chi_{ab}}{T} = -\frac{2A_s}{\varepsilon} \tag{6}$$

**Figure 2.** Experimental data of magnetic susceptibility for La$_{0.5}$Dy$_{0.5}$BaCaCu$_3$O$_{7-\delta}$.

**Figure 3.** Exemplification of experimental method used to find the reduced temperature $T_{C0}$ in the La$_{0.5}$Dy$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ sample.

**Figure 4.** Diamagnetism excess as a function of reduced temperature for the La$_{0.5}$Dy$_{0.5}$BaCaCu$_3$O$_{7-\delta}$ sample.


**Table I.** Critical parameters for the La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_7$–$\delta$ (Re = Y, Sm, Gd, Dy, Ho, Yb) superconductor.

<table>
<thead>
<tr>
<th>MUESTRA</th>
<th>$T_{c0}$ (K)</th>
<th>$T_C$ (K)</th>
<th>$B_{LD}(\times 10^{-2})$</th>
<th>$X$</th>
<th>$\lambda_4(\times 10^{-8})$</th>
<th>$\xi_{ab}(0)(\text{Å})$</th>
<th>$\xi_c(0)(\text{Å})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>La$<em>{0.5}$Y$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>74.00</td>
<td>80.02</td>
<td>0.58</td>
<td>-1.02</td>
<td>2.432</td>
<td>25.035</td>
<td>1.967</td>
</tr>
<tr>
<td>La$<em>{0.5}$Sm$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>58.00</td>
<td>80.01</td>
<td>0.53</td>
<td>-1.15</td>
<td>1.567</td>
<td>20.098</td>
<td>1.655</td>
</tr>
<tr>
<td>La$<em>{0.5}$Gd$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>74.72</td>
<td>80.22</td>
<td>0.57</td>
<td>-1.00</td>
<td>2.604</td>
<td>25.903</td>
<td>1.907</td>
</tr>
<tr>
<td>La$<em>{0.5}$Dy$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>54.96</td>
<td>77.05</td>
<td>0.52</td>
<td>-0.97</td>
<td>1.074</td>
<td>16.636</td>
<td>1.581</td>
</tr>
<tr>
<td>La$<em>{0.5}$Yb$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>46.37</td>
<td>66.14</td>
<td>0.58</td>
<td>-0.95</td>
<td>2.129</td>
<td>23.426</td>
<td>1.967</td>
</tr>
<tr>
<td>La$<em>{0.5}$Ho$</em>{0.5}$BaCaCu$_3$O$_7$–$\delta$</td>
<td>60.53</td>
<td>64.21</td>
<td>0.63</td>
<td>-1.02</td>
<td>2.564</td>
<td>25.707</td>
<td>2.358</td>
</tr>
</tbody>
</table>

In Eq. (6) it is clear that $\Delta Y/T$ varies as $\varepsilon^{-1}$, e.g., with an exponent $x=-1$. As observed in Table I, the exponent obtained by means of this adjustment for each one of the samples is consigned in the Table I, for all studied materials we obtain an $x=-1$. This result confirms that La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_7$–$\delta$ behaves as two-dimensional superconducting system.

4. **Conclusions**

La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_7$–$\delta$ (RE=Y, Sm, Gd, Dy, Ho, Yb) HTSC materials were synthesized by the solid state reaction technique. X-ray diffraction experiments shown that materials crystallized in a tetragonal perovskite structure (space group P4/mmm). From detailed susceptibility measurements we analyzed fluctuation effects close to critical temperature by means the Lawrence-Doniach model. In the limit of weak magnetic fields, results revealed that above and close the superconducting transition, La$_{0.5}$RE$_{0.5}$BaCaCu$_3$O$_7$–$\delta$ behaves as 2D anisotropic material. From analysis of diamagnetism excess we obtained values of critical parameters $B_{LD}$, $\xi_{ab}(0)$ and $\xi_c(0)$ for 2D fluctuation regime.

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