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Thermally activated phase-slip in high-temperature cuprates*

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Abstract

$\text{Gd}_{1-x-z}\text{Pr}_x\text{Ca}_z\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (GdPrCa-123) high-temperature superconductor cuprate (HTSC) samples with $0.0 \leq x \leq 0.3$ and $0.0 \leq z \leq 0.35$ were prepared by standard solid-state reaction and characterized by XRD technique. The magnetoresistance of the samples were measured and analysed. The superconducting transition region broadened by the application of a magnetic field. The resistive transition showed two distinct parts. (1) A steep part near the onset of superconductivity, where the onset transition temperature remained unchanged. For the $\text{Gd}_{0.85}\text{Ca}_{0.15}$ -123 sample, the steep part was more sensitive to the application of magnetic fields. (2) A transition tail part. The experimental data in this region were fitted with an Ambegakor and Halperin (AH) phase-slip model. The AH parameter, $\gamma(H)$, was used to estimate the critical current density at zero temperature of samples at various magnetic fields. It was observed that the critical current density decreased with Pr substitution and increased with Ca substitution in Gd-123. An optimal value of the Ca doping concentration for increasing the critical current density was determined. We suggest that the Pr ion probably acts to enhance the weak link and the Ca ion acts as a flux pinning centre in the GdPrCa-123 system. Moreover, we observed that the AH parameter, γ , and activation energy, U_0 , depend not only on temperature and magnetic field, but also on the Pr and Ca ion concentrations.

1. Introduction

After the discovery of high-temperature superconductor cuprates (HTSCs) by Bednorz and Muller, the magnetoresistance and broadening in the resistive transition of these new compounds, caused by both electrical current and magnetic field, have been topics of great interest and have been investigated extensively. In single crystals [1] and oriented thin films [2, 3] strong magnetic fields are required to show transition temperature broadening, while for granular materials small fields (~ 10 Oe) cause more broadening [4, 5]. Most authors such as Iwasaki *et al* [6], Palstra *et al* [7], Batlogg *et al* [8], Malozemoff *et al* [9], Gressin [10] and Koike *et al* [11] believe that the broadening behaviour in the low-resistance region near $T_c(\rho = 0)$ could be explained by a thermally activated flux creep model. However, observing the unusual broadening

in HTSC compounds, and also the broadening in resistive transition curves, even for a magnetic field applied parallel to the transitional current, leads many researchers to suggest different interpretations for the origin of this unusual observation. In this regard, reasons such as flux line melting [12], flux cutting, curved flux lines [13], flux entanglement [14] and thermally activated phase-slip [15] have been proposed.

In this paper, we have obtained interesting results by fitting the observed broadening in our samples to the Ambegakor–Halperin (AH) theory [15]. The AH theory describes effects of thermal fluctuations of the phase of the order parameter across a current-driven Josephson junction [16]. Near the transition temperature, thermal fluctuation can disrupt the coupling of the phase of the order parameter of the two superconductors forming a Josephson junction. In this theory, when $k_B T$ becomes comparable with the Josephson coupling energy, $\hbar I_c(T)/e$, where $I_c(T)$ is the maximum Josephson current at temperature T in the absence of noise, the resistance $\rho(T)$ in

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the limit of low bias current, $I \ll I_c(T)$, is given by

$$\rho(T) = \rho_n \left[I_0 \left(\frac{\gamma}{2} \right) \right]^{-2} \quad (1)$$

where I_0 is the modified Bessel function, ρ_n is the average normal state resistivity of the junction and γ is the normalized barrier height for thermally activated phase-slip (TAPS), defined as [15]

$$\gamma = \frac{U_0}{k_B T} = \frac{A(1-t)^q}{H}$$

where U_0 is the barrier height or the activation energy, H is the applied magnetic field, A is a constant and $t = T/T_c$. At temperatures near T_c , different values for the q power have been proposed: 0.85 [17], 1 [18], 1.5 [5, 19, 20], 1.2–1.25 [16], 2 [21, 22] and 1.53–2.8 [23]. Gaffney *et al* have suggested that the q power for granular YBCO is always less than 3/2 [17].

The AH model was first used to describe the unusual broadening observed in the resistance curves of YBCO crystals by Tinkham [19]; then in granular YBCO [16, 17, 21], Bi–Sr–Ca–Cu–O [5], and in the granular low-temperature superconductor NbN [24]. Good agreements between experimental and theoretical data have been observed through these investigations.

We have found a different way to obtain Pr and Ca ion effects in these high- T_c superconducting compounds by studying the magnetic properties of samples, and investigated their resistive transition curves under applied magnetic fields.

2. Experiment

The Pr- and Ca-substituted $\text{Gd}_{1-x-z}\text{Pr}_x\text{Ca}_z\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ cuprate superconductor samples with $0.0 \leq x \leq 0.3$ and $0.0 \leq z \leq 0.35$ have been prepared by a standard solid-state reaction. Stoichiometric amounts of 99.9% purity compounds, Gd_2O_3 , Pr_6O_{11} , CaCO_3 , BaCO_3 and CuO , were thoroughly mixed, ground and calcined in air atmosphere at 850 °C for 24 h. The calcination was repeated twice with intermediate grinding. The powder product was then pulverized and pressed into pellets. To obtain fully oxygenated samples, the pellets were annealed in the presence of an oxygen flow at 930 °C for 24 h on a gold sheet followed by slow cooling for 10 h to room temperature.

The samples were characterized at room temperature by powder x-ray diffraction using $\text{Cu K}\alpha_1$ radiation. The x-ray diffraction patterns were analysed using the Rietveld method. The oxygen content of the samples was then obtained using this analysis. The resistance and magnetoresistance measurements were carried out by a standard ac four-probe method over the temperature range 10–300 K. The ac current was 9.3 mA with a frequency of 70 Hz. The magnetic field was applied in the range of 0–2 T. The electrical resistance of the samples at room temperature was about 0.1 Ω . The temperature measurements were made with a Lake Shore-330 temperature controller with a Pt resistor and a GaAs diode. Dc magnetization measurements were performed using a vibrating sample magnetometer (VSM) with the sensitivity of 10^{-3} emu in a 2 T electromagnet.

3. Results and discussion

The x-ray analysis revealed that all the samples were single phase, having an orthorhombic structure with $Pmmm$ symmetry. The diffraction pattern analysis of compounds with high levels of Pr and Ca doping showed the presence of less than 1% impurity phases of BaCu_2O_2 and CaCu_2O_3 with the main peaks at $2\theta = 29.3^\circ$ and $2\theta = 33.94^\circ$, respectively. Using x-ray analysis, the average oxygen content of the samples was determined to be 6.87 ± 0.02 . The orthorhombic distortion, $((b-a)/(a+b)) \times 100$, decreased with increasing doping of Pr and Ca ions.

We investigated the resistive transition curves of ceramic compounds $\text{Gd}_{1-x-z}\text{Pr}_x\text{Ca}_z\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$. Figure 1 shows the resistive transition curves of Gd-123, $\text{Gd}_{0.85}\text{Ca}_{0.15}$ -123, $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}$ -123 and $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}$ -123 in a magnetic field applied perpendicular to the direction of the electrical current. A broadening of the transition was observed in the resistive curves.

As can be seen in figure 1, the resistive transition shows two distinct parts: a steep part ($\Delta T \approx 1$ K) in the onset of the superconductivity transition region, where the onset transition temperature remains unchanged under different magnetic fields, and a tailing part. The steep region was less sensitive to the applied magnetic fields. For the $\text{Gd}_{0.85}\text{Ca}_{0.15}$ -123 sample the steep part was more sensitive to the magnetic field, showing a larger broadening on increasing the field. The reason behind this difference is under investigation. A much broader tail followed the steep part. The tail part was associated with weak links between the grain [16]. This region is extremely sensitive to the magnetic field and moves to lower temperatures on increasing the magnetic field. This sensitivity decreases on increasing the field (the magnetoresistance curves move slowly to lower temperatures). The experimental data in this region were fitted to the AH phase-slip model with four fitting parameters. To fit the data, we assumed $\gamma = C(1-t)^q$, where C is a magnetic-field-dependent parameter. So, we could determine the γ variation with respect to temperature, magnetic field and doping concentrations of Pr and Ca ions.

Figure 2 shows the experimental magnetoresistance data and the fitted curves to the AH model for Gd-123, $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}$ -123 and $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}$ -123 samples. For the fitting of the data of each compound we chose four free fitting parameters: ρ_n , C , T_c and q . The value of ρ_n was observed to be constant with increasing applied magnetic field; it was close to the resistance value of the branching point of the magnetoresistance curves in figure 1. We found a minimum for ρ_n , equal to $\sim 114 \mu\Omega \text{ cm}$ at $T_c = 95.7$ K for Gd-123, and a maximum for ρ_n , equal to $\sim 338 \mu\Omega \text{ cm}$ at $T_c = 90.5$ K for $\text{Gd}_{0.85}\text{Ca}_{0.15}$ -123. Moreover, with increasing Ca ion concentration in $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}$ -123 ρ_n decreases. The obtained value is $\sim 300 \mu\Omega \text{ cm}$ at $T_c = 83$ K for $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}$ -123 and $\sim 180 \mu\Omega \text{ cm}$ at $T_c = 80$ K for $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}$ -123. The ρ_n variation shows that it increases by adding Pr and Ca ions to Gd-123. The reason is probably the distance of intergranular links, which increases the system's resistivity. In the fitting process, the obtained T_c value is close to the superconducting onset temperature, which remains almost unaffected under the applied field. Of course, for $\text{Gd}_{0.85}\text{Ca}_{0.15}$ -123 the obtained T_c is more than the onset temperature of superconductivity and

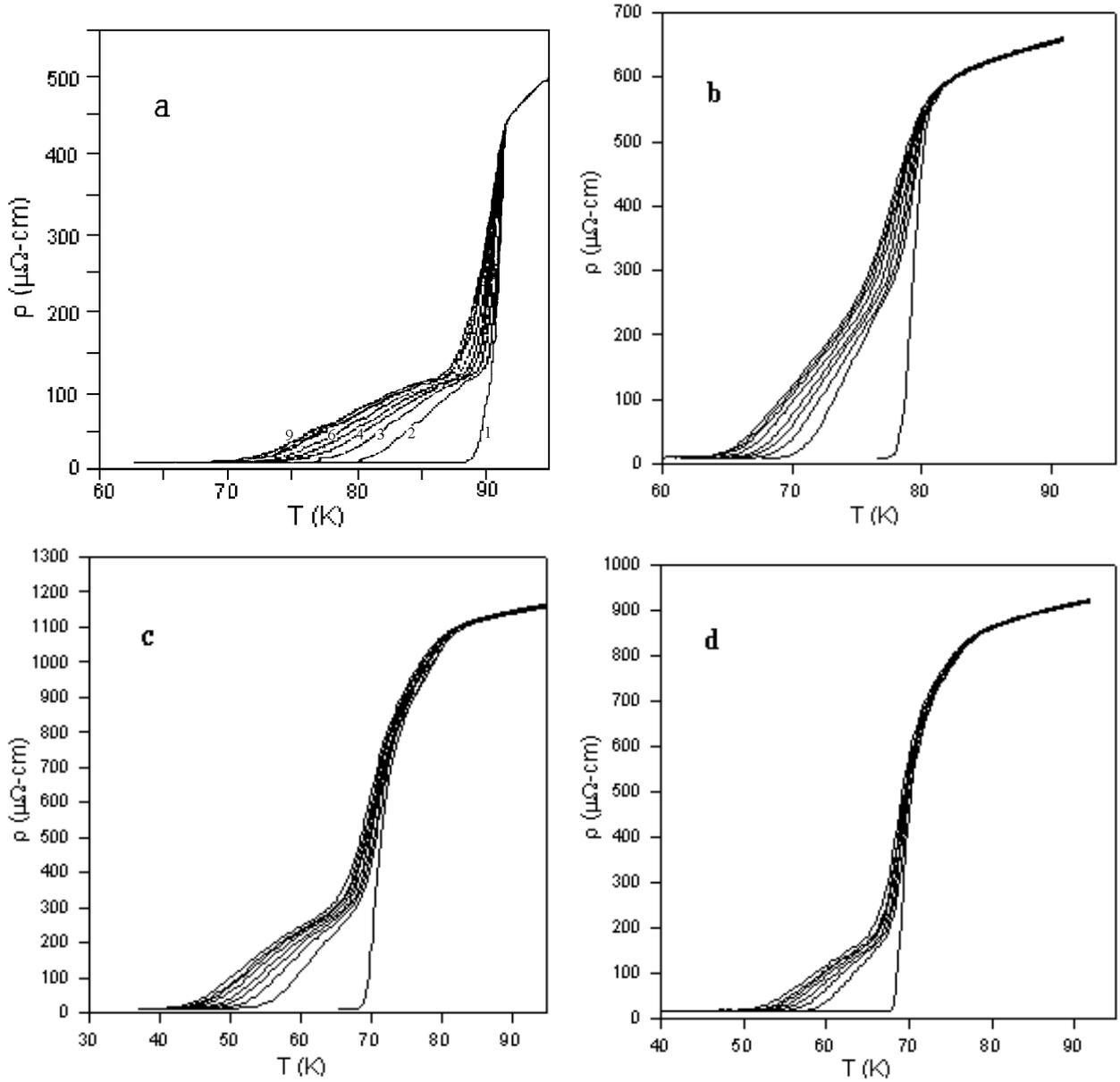


Figure 1. Resistive transition as a function of temperature at various magnetic fields for (a) Gd-123, (b) $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$, (c) $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}\text{-123}$ and (d) $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$ compounds. The magnetic fields are: 1, 0.0; 2, 1.0; 3, 3.0; 4, 5.0; 5, 7.0; 6, 10.0; 7, 13.0; 8, 15.0 and 9, 17.0 kOe.

for $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$ T_c is close to the mid-temperature of the superconducting transition (the temperature at which the resistance is half that of the extrapolated normal state). Moreover, it is shown that with increasing applied magnetic field, q increases and parameter C decreases, in agreement with [16]. As a result, the temperature dependence of γ for Gd-123 moves from $(1-t)^{1.57}$ for $H = 1.0$ kOe to $(1-t)^{2.4}$ for $H = 15.0$ kOe. The q exponent value is equal to $q = 1.57\text{--}2.4$, $q = 1.78\text{--}2.91$, $q = 2.80\text{--}2.87$ and $q = 2.84\text{--}3.05$ for Gd-123, $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$, $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}\text{-123}$ and $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$, respectively. At temperatures below the transition temperature, the increase of q and the decrease of C result in a decrease in the activation energy, U_0 , in equation (1).

In the next step of the fitting procedure we chose the value of ρ_n to be constant and equal to $110 \mu\Omega \text{ cm}$ for Gd-123 (obtained using figure 1). The other three parameters were

taken to be variables. It is observed that on increasing the field, q increases and C decreases. The average of q is 2.33. Then, we took $q = 2.33$ and $\rho_n = 110 \mu\Omega \text{ cm}$, and the other two parameters were taken as variables. We observed that the value of T_c is nearly constant, close to T_c onset, and the variation of C is the same as before. The values obtained for the $C(H)$ parameter are shown in table 1. For the $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$ sample, due to the large broadening of the steep part of the magnetoresistance curve, and unknown ρ_n , we have not designated a constant value to ρ_n . In this compound, however, we chose $q = 2.91$ (the average q of the previous fitting) and the other parameters as free. We obtained reasonable results for T_c , ρ_n and C (table 1). In the next step, we chose ρ_n as constant, equal to $319 \mu\Omega \text{ cm}$ and $200 \mu\Omega \text{ cm}$ (obtained from figure 1) for the $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}\text{-123}$ and $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$ compounds, respectively. It was seen that the obtained T_c

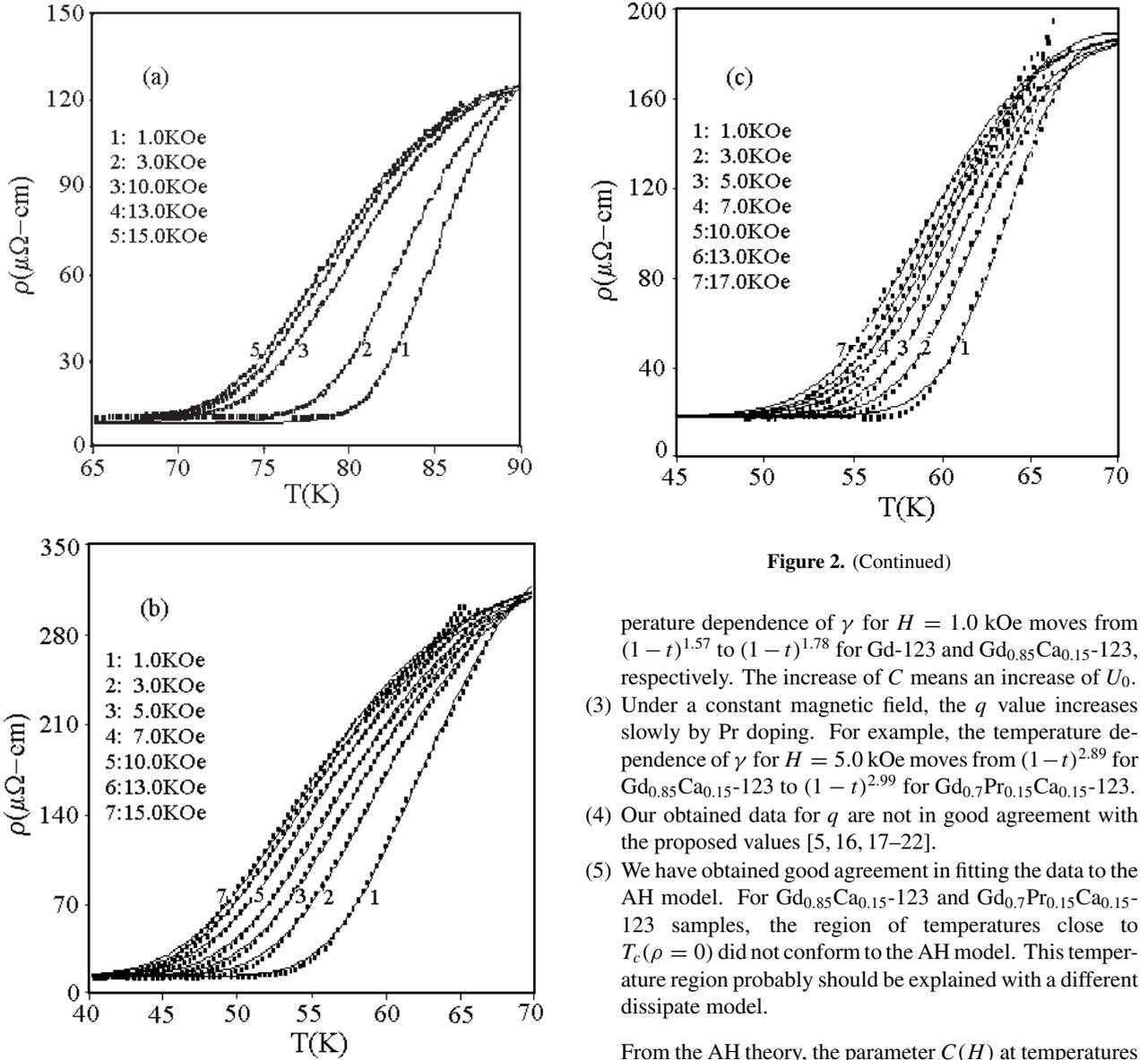


Figure 2. (Continued)

perature dependence of γ for $H = 1.0$ kOe moves from $(1-t)^{1.57}$ to $(1-t)^{1.78}$ for Gd-123 and $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$, respectively. The increase of C means an increase of U_0 .

- (3) Under a constant magnetic field, the q value increases slowly by Pr doping. For example, the temperature dependence of γ for $H = 5.0$ kOe moves from $(1-t)^{2.89}$ for $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$ to $(1-t)^{2.99}$ for $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$.
- (4) Our obtained data for q are not in good agreement with the proposed values [5, 16, 17–22].
- (5) We have obtained good agreement in fitting the data to the AH model. For $\text{Gd}_{0.85}\text{Ca}_{0.15}\text{-123}$ and $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$ samples, the region of temperatures close to $T_c(\rho = 0)$ did not conform to the AH model. This temperature region probably should be explained with a different dissipate model.

From the AH theory, the parameter $C(H)$ at temperatures close to the transition temperature is given by [16]

$$C = \frac{J_{cj}(0)\hbar a^2}{ek_B T_c} \quad (2)$$

where $J_{cj}(0)$ is the critical current density at zero temperature and a is the average grain size. The average grain size of the samples obtained by SEM is about $1 \mu\text{m}$. Using the values of $C(H)$ (table 1) and equation (2), we estimate the critical current density at zero temperature for the samples in table 2. Reviewing table 2 we can see that on increasing the magnetic field, the critical current density of samples decreases, in agreement with [16]. An increase in the magnetic field results in the penetration of more magnetic flux lines in the sample and an increase in the Lorentz force due to the field increment causes more flux lines to move. Thus, it is concluded that the critical current density decreases accordingly.

When a Ca impurity was added to Gd-123, $J_{cj}(0)$ increased; also, when the Ca impurity was added, up to the optimum doping limit, to GdPr-123 at $H = 1.0$ kOe, $J_{cj}(0)$ increased from $19.9 \times 10^3 \text{ A cm}^{-2}$ for $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}\text{-123}$

Figure 2. Resistance transition as a function of temperature under various magnetic fields, truncated at the onset of broadening, for (a) Gd-123, (b) $\text{Gd}_{0.75}\text{Pr}_{0.15}\text{Ca}_{0.1}\text{-123}$ and (c) $\text{Gd}_{0.7}\text{Pr}_{0.15}\text{Ca}_{0.15}\text{-123}$ compounds. The full curves show the fits to equation (1).

and q parameters were irrational. Then, we chose $q = 2.80$ and $q = 2.95$ (the average of q in the previous fitting) for $\text{GdPr}_{0.15}\text{Ca}_{0.1}\text{-123}$ and $\text{GdPr}_{0.15}\text{Ca}_{0.15}\text{-123}$, respectively, and the other parameters as free. The obtained results are reasonable and coincide with the previous results.

During the fitting process, it was noticed that q and T_c were related to each other: for example if one was constant, the other one became constant too.

With due attention to these and table 1, we can summarize the following results:

- (1) On increasing the magnetic field for each compound, the q values increase and the C parameter decreases. These variations result in a decrease of the activation energy U_0 .
- (2) Under a constant applied field, the q and the C parameters increase by Ca doping to Gd-123. For example, the tem-

Table 1. Values of the parameter $C(H)$ for granular GdPrCa-123 compounds.

Sample	$H(\text{kOe})$							
	1	3	5	7	10	13	15	17
Gd-123	360	250	210	188	168	143	138.3	—
Gd _{0.85} Ca _{0.15} -123	460	340	300	309	244	—	230	210
Gd _{0.75} Pr _{0.15} Ca _{0.1} -123	114	76.1	74	57.8	59.5	51.8	54.3	—
Gd _{0.7} Pr _{0.15} Ca _{0.15} -123	320	220	176	150	130	124.4	101	116.3

Table 2. The critical current density at zero temperature, $J_{cj}(0) \times 10^3 \text{ A cm}^{-2}$, for granular GdPrCa-123.

Sample	$H(\text{kOe})$							
	1	3	5	7	10	13	15	17
Gd-123	72.0	49.6	41.7	37.5	33.6	28.6	27.7	—
Gd _{0.85} Ca _{0.15} -123	87.7	65.2	57.6	58.2	46.4	—	43.6	40.3
Gd _{0.75} Pr _{0.15} Ca _{0.1} -123	19.9	13.4	12.8	10.3	10.1	8.98	9.2	—
Gd _{0.7} Pr _{0.15} Ca _{0.15} -123	52.2	35.9	28.7	24.4	21.7	20.8	17.1	19.6

to $52.16 \times 10^3 \text{ A cm}^{-2}$ for Gd_{0.7}Pr_{0.15}Ca_{0.15}-123. We suggest that the Ca ion acts as a flux pinning centre in agreement with [25], which results in an increase in the critical current density. Observing the impurity phases in the x-ray diffraction pattern for high values of Ca doping, one concludes that it is probable that the critical current density of the system does not increase with Ca addition for more than the optimum value. Hence, the excess Ca does not act as pinning centres.

A more detailed examination of table 2 tells us that the value of $J_{cj}(0)$ decreases by Pr doping into GdCa-123, for example $J_{cj}(0)$ decreases from $88 \times 10^3 \text{ A cm}^{-2}$ for GdCa_{0.15}-123 to $52 \times 10^3 \text{ A cm}^{-2}$ for GdPr_{0.15}Ca_{0.15}-123 under $H = 1.0 \text{ kOe}$. We suggest that the Pr ion probably acts as a weak link, which causes the critical current density to decrease.

Figure 3 shows the variation of parameter γ as a function of magnetic field for GdPrCa-123. It can be seen that on increasing the Ca ion content in the samples, the $\gamma(H)$ parameter increases, and on increasing the Pr ion content it decreases. Using figure 3, we obtain the magnetic field dependence of γ to be of the form $\gamma(H) \propto H^{-n}$. We also obtained $n = 0.36, 0.26, 0.28$ and 0.39 for Gd-123, Gd_{0.85}Ca_{0.15}-123, Gd_{0.75}Pr_{0.15}Ca_{0.1}-123 and Gd_{0.7}Pr_{0.15}Ca_{0.15}-123, respectively.

4. Conclusions

The broadening of the resistive transition observed for GdPrCa-123 in the presence of an external magnetic field was attributed to the phase-slip phenomenon due to thermally activated motion of vortices according to the AH model. The magnetic field is thought to cause the phase-slip to increase between the two superconductors which make the Josephson junction, and, consequently, the increase in the applied magnetic field increases the phase-slip rate and broadens the resistive transition. We found that the exponent q in the temperature dependence of the parameter γ varies with respect to the increase of the magnetic field and Pr and Ca concentrations. We observed that the γ parameter depends not only on the temperature and magnetic field, but also on the Pr and Ca concentrations in the system. Under a constant applied magnetic field, the γ parameter or the activation energy, U_0 ,

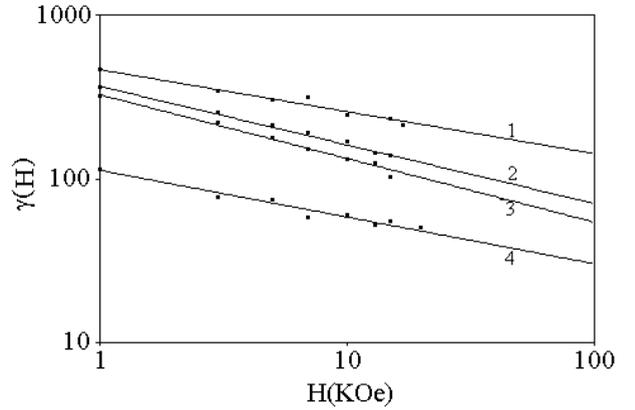


Figure 3. The magnetic field dependence of the normalized activation energy, γ , as obtained from the magnetoresistivity data for 1, GdCa-123; 2, Gd-123; 3, Gd_{0.75}Pr_{0.15}Ca_{0.1}-123; and 4, Gd_{0.7}Pr_{0.15}Ca_{0.15}-123. The full curves show the result of the fit to the AH model.

increases by Ca doping, and the temperature dependence of γ varies due to Pr and Ca dopings in the system.

We have estimated the critical current density at zero temperature, $J_{cj}(0)$, for the samples using the parameter γ . It is observed that in GdPrCa-123 compounds at an optimum value for Ca doping the Ca ions act as flux pinning centres, which results in the increase in the critical current density. On the other hand, Pr doping led to a low critical current density relative to the GdCa-123 case. We suggest that the Pr ions probably act as weak links. Using the magnetic field dependence of the γ parameter to be of the form $\gamma(H) \propto H^{-n}$, we obtained $n = 0.36, 0.26, 0.28$ and 0.39 for Gd-123, Gd_{0.85}Ca_{0.15}-123, Gd_{0.75}Pr_{0.15}Ca_{0.1}-123 and Gd_{0.7}Pr_{0.15}Ca_{0.15}-123 samples, respectively.

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