Journal of Low Temperature Physics, Vol 117, Nos 5/6, 1999

Percolation Analyses on Transport Properties of $YBa_2Cu_3O_{7-\delta}$-Ag System

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The transport properties of $YBa_2Cu_3O_{7-\delta}$-Ag composite ceramic system have been studied from the viewpoint of the percolation theory. The percolation threshold volume fraction f_c of Ag determined from the electrical resistivity and from the thermal conductivity is f_c=0.125±0.005. This markedly smaller value than the theoretical value (f_c=0.16) suggests the segregative distribution of Ag. The critical exponent t depends on the component conductivity ratio $h=\sigma_{YBCO}/\sigma_{Ag}$, which can be explained on the basis of the scaling hypothesis.

PACS number: 72.15.Cz, 71.30.+h

1. INTRODUCTION

One main hurdle against the application of high T_c oxide superconductors in ceramic forms is the occurrence of the weak-links at the border of grains, which practically limit the effective transport critical current I_c^{eff} of bulk cuprate superconductors. The addition of Ag metal to cuprates is often effectual to make up for the weak-links. It is physically interesting and also useful for an applicational purpose to investigate the behavior of Ag clusters on the basis of the percolation theory. The accumulated data and analyses from the viewpoint of the percolation theory, however, are somewhat scattered and sometimes inconsistent with each other.[1-4]

In this study, we try to develop synthetic analyses about the percolative behavior of Ag clusters in $YBa_2Cu_3O_{7-\delta}$ (YBCO) by measuring the electrical resistivity and the thermal conductivity. The conductivity ratio of Ag metal to the cuprate is widely changed by varying the amount of oxygen deficiency δ of YBCO. The experimental results are analyzed on the basis of the scaling theory for the percolation of Ag clusters.

2. OUTLINE OF THE SCALING LAW

In a two component random composite of *M* and *I*, clusters of *M* with an infinite scale are formed when the volume fraction f_M of *M* exceeds a certain threshold value f_c. Assuming *M* to be a metal and *I* to be an insulator, a power law holds for the bulk effective conductivity σ_E of the composite in the critical region;

$$\sigma_E = A\sigma_M(f_M - f_c)^t \quad \text{for } 0 < f_M - f_c << 1. \tag{1}$$

The values of $t = 1.9 \pm 0.1$ and $f_c \approx 0.16$ were predicted for three-dimensional continuum media.[5] If *I* component is also somewhat conductive and the conductivity ratio $h = \sigma_I/\sigma_M$ is not strictly zero, the power law of eq. (1) breaks down as $\Delta f = f_M - f_c$ tends to zero. In this case, σ_E is expected to follow the scaling hypothesis,[6]

$$\sigma_E = \sigma_M \Delta f^t \Phi_+(z), \tag{2}$$

where $z = h/\Delta f^{t+s}$, $s(\approx 0.73)$ is the critical exponent governing the divergence of the conductivity in a superconductor-metal composite for $0 < f_c - f_M << 1$. The universal scaling function $\Phi(z)$ can be expanded in the power series of z:

$$\Phi_+(z) = A^+ + B^+ z + C^+ z^2 + \cdot\cdot\cdot \quad \text{for } f_M - f_c > 0 \text{ and } z < 1. \tag{3}$$

For non-zero values of h, the conductivity data must be carefully handled taking proper account of region of z in which the application of eq. (3) is justified. In this paper, h dependence of the t exponent is mainly discussed.[4,7]

3. EXPERIMENTAL

YBCO powder and Ag_2O powder (Dowa Mining Co. Ltd.) were mixed with an appropriate weight ratio and pressed into pellets. The pellets were pre-sintered at 935°C for 15h in air and furnace cooled. The pre-sintered pellets were crushed, mixed and again pressed into pellets. They were sintered at 930°C for 30h in air and then furnace cooled in 15h. After the obtained pellets were cut into square pillars (~3x3x15mm^3), they were heated again and quenched into liquid N_2 from various temperatures to prepare 'quenched samples'.

The density of the samples was determined by measuring the size and the mass. Figure 1 shows the filling factor of the samples, which increases with increasing nominal Ag volume fraction $f_{Ag}{}^N$. In this paper, the Ag volume fraction is corrected using the least square fitting curve in Fig. 1. The electrical resistivity and the thermal conductivity were automatically measured between 10K and 260K in a Gifford-McMahon refrigerator.[8]

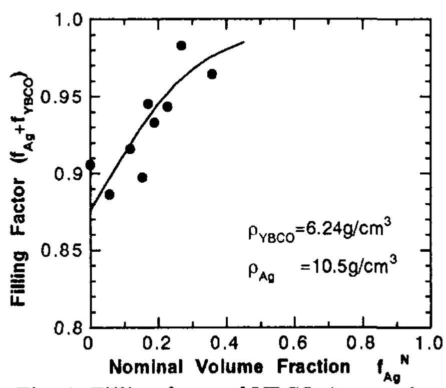

Fig. 1. Filling factor of YBCO-Ag samples as a function of nominal Ag volume fraction f_{Ag}^N. f_{Ag}^N was corrected using the least square fitting line to give f_{Ag}.

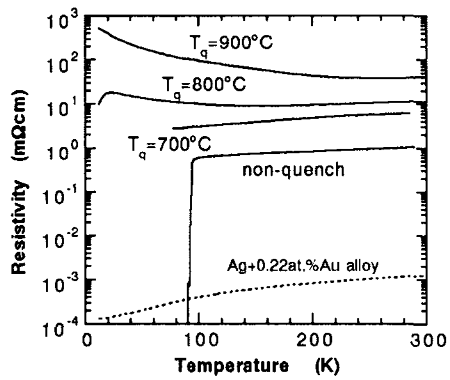

Fig. 2. Temperature dependence of the electrical resistivity $\rho(T)$ of YBCO non-quenched and quenched from 700°C, 800°C and 900°C.

4. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of the electrical resistivity $\rho(T)$ of YBCO non-quenched, and quenched from 700°C, 800°C and 900°C. $\rho(T)$ of YBCO rapidly increases with increasing quenching temperature T_q. In the figure, $\rho(T)$ of Ag+0.22at.%Au alloy whose residual resistivity ratio (RRR) is nearly equal to the estimated RRR of Ag clusters in YBCO[9] is also shown.

Figures 3(a)-3(c) show typical plots of log σ_E vs log $(f_{Ag}-f_c)$ to determine the experimental values of critical exponent t_{ex} and the percolation threshold f_c. The conductivity values at 150K were used because RRR of Ag cluster is verified to depend on f_{Ag} to some extent[9] and the $\sigma_{YBCO}/\sigma_{Ag}$ values become dependent on RRR of Ag cluster at low temperatures. The most suitable value of f_c was determined to be 0.125 for the present Ag+YBCO ceramics system. This f_c value is remarkably smaller than the generally accepted value of $f_c \approx 0.16$ for 3D continuum media. The smaller value of observed f_c should reflect selective distribution of Ag clusters into voids at grain boundaries of YBCO; the filling factor in Fig. 1 rapidly increases with increasing Ag composition, which also suggests that Ag metals fill up the voids of ceramic YBCO preferentially.

In order to determine t_{ex}, the region of $\Delta f(=f_{Ag}-f_c)$ must be carefully set up. Eqs.(2) and (3) are valid for $\Delta f>(\sigma_{YBCO}/\sigma_{Ag})^{1/s+t}$ which corresponds to the condition, $z<1$. On the other hand, if we go too far from f_c, we pass through the critical region and enter into the region where the effective medium approximation (EMA) becomes valid. In this work, we set the region to determine t_{ex} as $(\sigma_{YBCO}/\sigma_{Ag})^{1/s+t} \leq \Delta f \leq 0.2$. The determined t_{ex} values depend on the conductivity ratio h and are larger for smaller h as shown in Fig. 4. In the following, we estimate the relation between observed t_{ex} value and h on the basis of the scaling law given by eqs. (2) and (3).[4,6,7]

To calculate σ_E based on eq. (2), the scaling function Φ_+ of eq. (3) must be known. As a trial function, we set $\Phi_+(z)=A^+(1+z)$ and calculated σ_E in the range, $(\sigma_{YBCO}/\sigma_{Ag})^{1/s+t} \leq \Delta f \leq 0.2$. Then average slope between $\Delta f=(\sigma_{YBCO}/\sigma_{Ag})^{1/s+t}$ and $\Delta f=0.2$ was defined as t^* and obtained t^* is given by the dotted line in Fig. 4. In the present calculation, the t value of 1.8 and s value of 0.73 were used. The t^* values

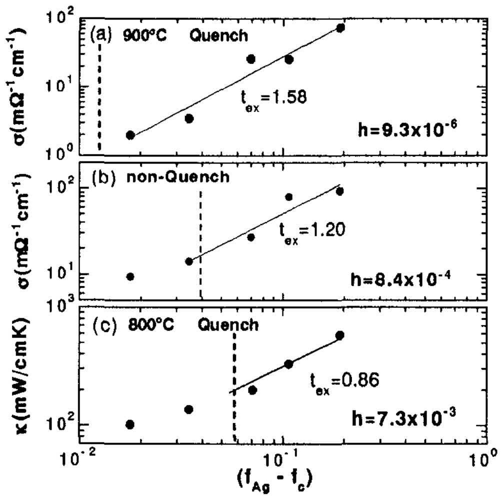

Fig. 3. The typical plots of log σ_E vs log $(f_{Ag}-f_c)$ to determine experimental values of critical exponent t_{ex} and the percolation threshold f_c for (a) T_q=900°C, (b) non-quenched and (c) T_q=800°C, respectively. Note that (c) is for the thermal conductivity. The vertical dashed line represents Δf value for respective samples which corresponds to z=1.

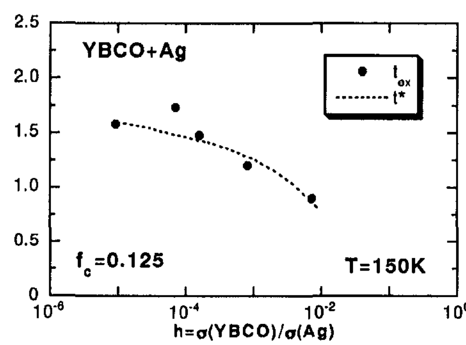

Fig. 4. The experimentally determined t_{ex} values and calculated t^* values as a function of conductivity ratio h=σ(YBCO)/σ(Ag) (see text).

Table I. The parameter values used for the calculation of t^* and experimental t_{ex} values. For the $\rho(T)$ analyses, $\sigma_{Ag}=1621 m\Omega^{-1}cm^{-1}$ was used from Fig. 2.

measurement	T_q	conductivity at 150K	$h(=\sigma_{YBCO}/\sigma_{Ag})$	t_{ex}	t^*
κ	800°C	κ_{Ag}=4300mW/cmK κ_{YBCO}=31.3mW/cmK	7.3×10^{-3}	0.86	0.78
ρ	non-Q	$\sigma_{YBCO}=1.36\ m\Omega^{-1}cm^{-1}$	8.4×10^{-4}	1.20	1.27
ρ	700°C	$\sigma_{YBCO}=0.26\ m\Omega^{-1}cm^{-1}$	1.6×10^{-4}	1.48	1.43
ρ	800°C	$\sigma_{YBCO}=0.114 m\Omega^{-1}cm^{-1}$	7.0×10^{-5}	1.73	1.49
ρ	900°C	$\sigma_{YBCO}=0.015 m\Omega^{-1}cm^{-1}$	9.3×10^{-6}	1.58	1.99

in Fig. 4 agree fairly well with the experimental t_{ex} values. The parameter values used for the calculation are summarized in Table I.

5. SUMMARY

(1) The electrical resistivity ρ and the thermal conductivity κ were measured for $Ag+YBa_2Cu_3O_{7-\delta}$ composites and the results were analyzed. The percolation threshold volume fraction f_c of Ag is determined to be 0.125, which is remarkably smaller than the generally accepted value of 0.16 for random binary mixing in three-dimensional continuum media. The observed small f_c value supports the segregative distribution of Ag particles in this system.
(2) Observed critical exponent t_{ex} for $f_{Ag}>f_c$ is dependent on the conductivity ratio $h=\sigma(YBa_2Cu_3O_{7-\delta})/\sigma(Ag)$, which was varied in this study by quenching the composites from various temperatures.
(3) On the basis of the scaling law, the trial scaling function, $\Phi_+(z)=A(1+z)$, explains fairly well the h dependence of the observed t_{ex} values.

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