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Model analyses of thermal conductivity and purity of doped Ag in $\mathbf{Ag + YBa_2Cu_3O_7}$

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The thermal conductivities $\kappa(T)$ of Ag-doped $YBa_2Cu_3O_7$ sintered materials were measured for various Ag fractions up to $f_{Ag} = 0.67$. By analyzing the $\kappa(T)$ data based on a proposed phenomenological model, the purity of doped Ag in $YBa_2Cu_3O_7$ could be determined.

Key words: thermal conductivity, Ag doped $YBa_2CU_3O_7$, purity of Ag.

1. Introduction

Ag-doping of sintered high-T_c cuprates is often effective in improving superconducting characteristics, reinforcing the weak links between the grain boundaries. The transport properties, such as the thermal conductivity $\kappa(T)$ and the electrical resistivity $\rho(T)$, of Ag-doped materials are heavily influenced by the content and the purity of the doped Ag. κ is an important parameter which dictates the dynamic stability of a superconducting material. We previously studied the Ag distribution and the Ag purity in Ag-doped Bi2212 crystals fabricated by a floating zone (FZ) method and proposed a simple phenomenological model to analyze the thermal and electrical transport in this composite system [1, 2]. The crystal grains and Ag distribution were highly anisotropic in the FZ crystals.

In this paper, we report the temperature dependence of $\kappa(T)$ of Ag-doped $YBa_2Cu_3O_7$ (YBCO) sintered materials with Ag volume fractions up to $f_{Ag} = 0.67$. We apply the phenomenological model to analyze the $\kappa(T)$ data of Ag + YBCO composite materials.

2. Experimental

The Ag-doped YBCO samples were fabricated as follows; YBCO powder and Ag_2O powder (Dowa Mining Co. Ltd) were mixed with an appropriate weight ratio and pressed into pellets. The average sizes of each powder were about 7 μm and 10 μm, respectively. After they were sintered at 550 $\sim$ 880 °C for 8 hours in air, they were pulverized, again pressed into pellets and sintered at 925 $\sim$ 935 °C for 30 hours in air followed by furnace cooling. The Ag volume fraction f_{Ag} was calculated from ideal densities $d_{Ag} = 10.25$ and $d_{YBCO} = 6.24$ and the initial weight ratio w_{Ag}/w_{YBCO}. The thermal conductivity $\kappa(T)$ was measured from 10 to 200 K using an automated measuring system which employed a GM refrigerator as a cryostat [3].

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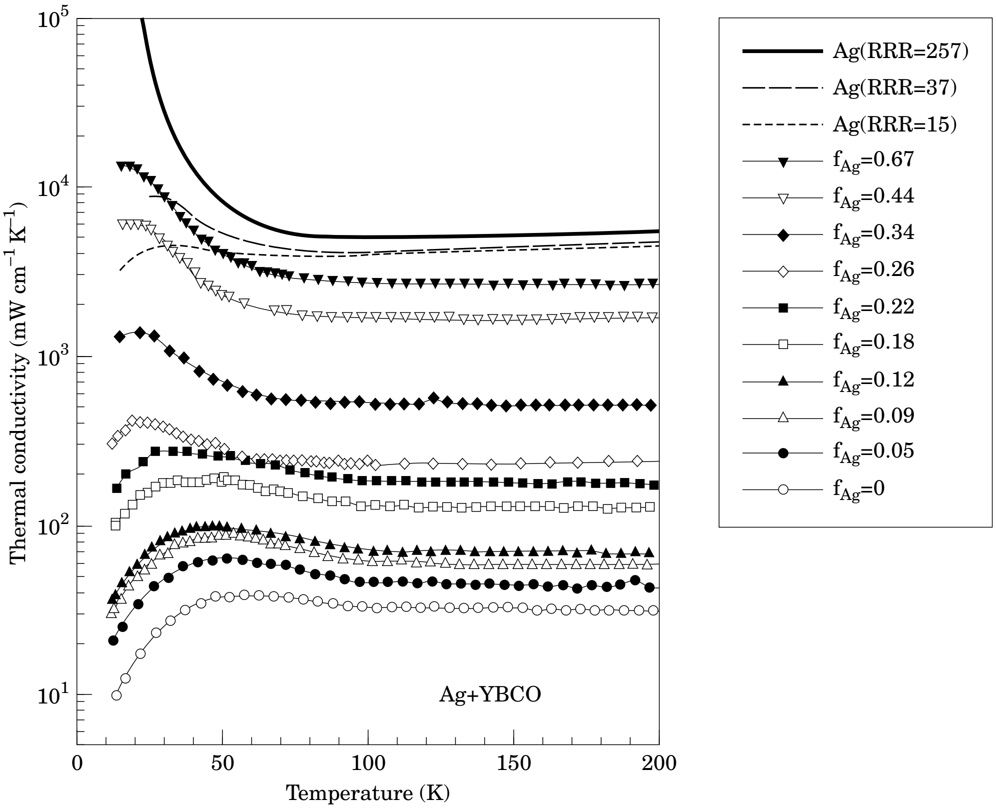

Fig. 1. The temperature dependence of the thermal conductivity $\kappa(T)$ of Ag + YBCO materials with various Ag concentrations f_{Ag}, together with that of Ag-Au alloys [5].

3. Results and discussion

It was confirmed from the analyses by an electron probe microanalyzer (EPMA) that the doped $\mathrm{Ag_2O}$ powder was uniformly and isotropically dispersed in the form of metal particles and that the content of Ag particles increased with increasing nominal $\mathrm{Ag_2O}$ concentration [4]. The $\rho(T)$ values of Ag-doped YBCO decreased with increasing Ag content, and the T_{c} of about 92 K was observed up to $f_{\mathrm{Ag}} = 0.34$. The κ values increased with increasing Ag content as shown in Fig. 1. In Ag + YBCO composite materials, the heat should flow through a YBCO path, a Ag path or series of YBCO and Ag paths. Then the thermal resistance of Ag + YBCO composites is given by the general equivalent circuit in Fig. 2A. Taking account of the isotropic nature of this sintered material, we propose a simplified model cube of Ag + YBCO specimen in Fig. 2B, which exemplifies the equivalent circuit of Fig. 2A [1, 2]. In this model the doped Ag particles are divided into two groups. One group consists of linked Ag particles, presumably situated mainly in the grain boundaries, which form Ag pillars and contribute to a parallel heat path. The other group consists of isolated Ag particles which form a smaller inside cube and contribute to the series path of Ag and YBCO. The sum of volumes of pillars and a cube is postulated to be equal to f_{Ag}. The total thermal resistance W of the circuit in Fig. 2A is given by

$$\frac{1}{W} = \frac{1}{W_{\mathrm{Y1}}} + \frac{1}{W_{\mathrm{Ag1}}} + \frac{1}{W_{\mathrm{Y2}} + W_{\mathrm{Ag2}}} + \frac{1}{W_{\mathrm{Y3}} + W_{\mathrm{Ag3}}}. \tag{1}$$

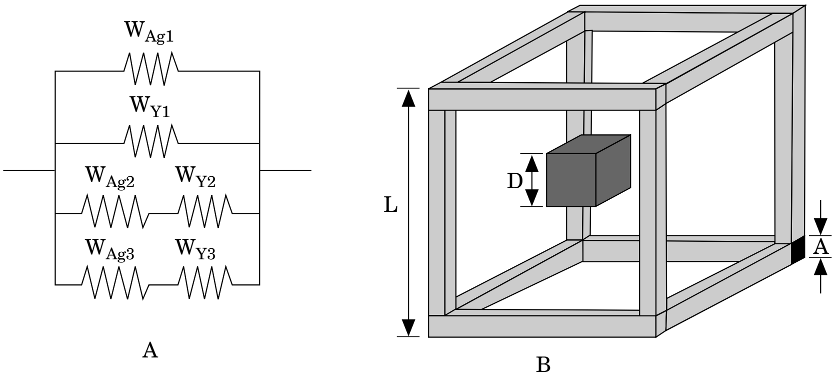

Fig. 2. A, A general equivalent circuit of the thermal resistance of Ag + YBCO composites. B, A simplified model cube of the composite which takes into account the isotropic nature of the Ag distribution in this sintered material.

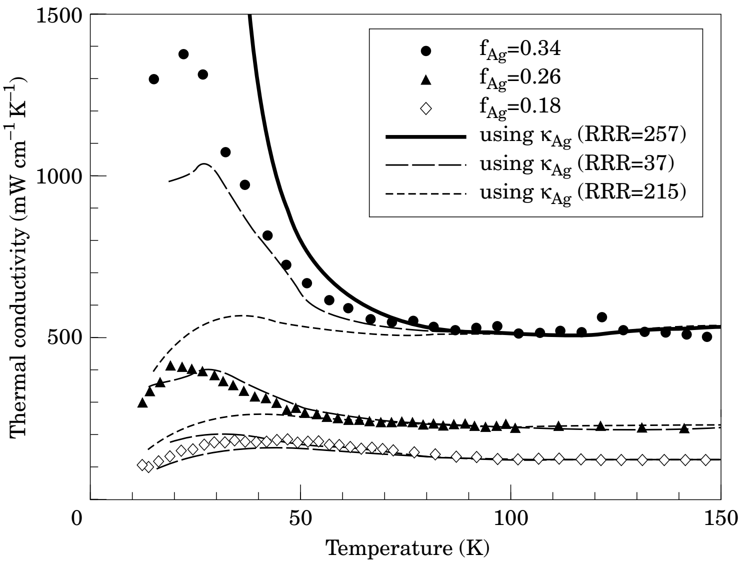

Fig. 3. Examples of the comparison of the measured $\kappa(T)$ with the calculated $\kappa(T)$ of Ag + YBCO composite materials using $\kappa_{\mathrm{Ag}}(T)$ of $RRR = 257$, 37 and 15.

If one edge of the model cube has length L, if the edge of the inner cube is D and if the cross-section of a pillar is A, each thermal resistance component is approximated as $W_{\mathrm{Y1}} = L/\{\kappa_{\mathrm{YBCO}}(L - D - 2A)(L + D - 2A)\}$, $W_{\mathrm{Y2}} = (L - D)/(\kappa_{\mathrm{YBCO}} D^2)$, $W_{\mathrm{Y3}} = 1/(4\kappa_{\mathrm{YBCO}} A)$, $W_{\mathrm{Ag1}} = L/(4\kappa_{\mathrm{Ag}} A^2)$, $W_{\mathrm{Ag2}} = 1/(\kappa_{\mathrm{Ag}} D)$ and $W_{\mathrm{Ag3}} = 1/\{2\kappa_{\mathrm{Ag}}(L - 2A)\}$ with κ_{YBCO} and κ_{Ag} being the thermal conductivities of YBCO and Ag, respectively. For κ_{YBCO} we used the value for an undoped YBCO ($f_{\mathrm{Ag}} = 0$) sample, and for κ_{Ag} were used those of Ag + Au alloys [5] (residual resistivity ratio (RRR): $\rho(280\ \mathrm{K})/\rho(4.2\ \mathrm{K}) = 257$, 37 and 15) which are also shown in Fig. 1. A and D were determined so as to achieve the best fit to be observed $\kappa(T)$ values at three temperatures of 80 K, 100 K and 150 K. Figure 3 shows examples of the fitting. The temperature dependence of $\kappa(T)$ was well reproduced by the model.

As is well-known, $\kappa(T)$ of a pure metal only weakly depends on its purity in the high temperature region

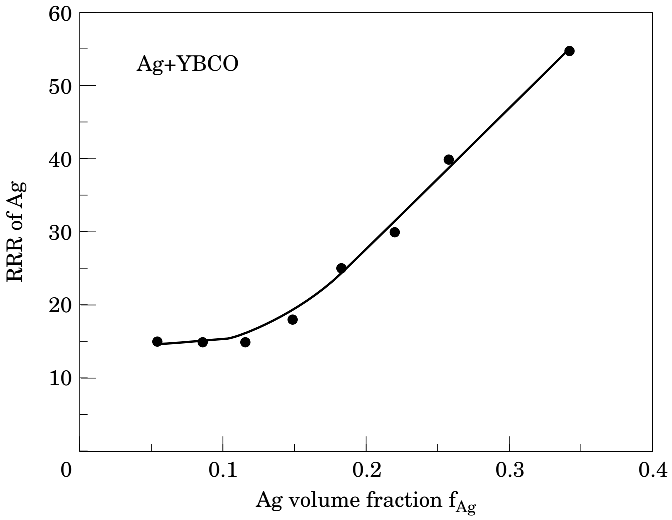

Fig. 4. Estimated RRR of Ag in Ag + YBCO versus Ag volume fraction f_{Ag}.

($T > 70$ K for Ag). In the low temperature region, however, $\kappa(T)$ is very sensitive to purity and is strongly enhanced in purer metals. For the sample of $f_{\text{Ag}} = 0.18$, the experimental $\kappa(T)$ is between the calculated $\kappa(T)$ using κ_{Ag} of $RRR = 15$ and that using $RRR = 37$, which means that the purity of doped Ag is between $RRR = 15$ and 37. It was confirmed that the doped-Ag purity in the sample $f_{\text{Ag}} = 0.26$ was nearly equal to $RRR = 37$ and that in $f_{\text{Ag}} = 0.34$ was between $RRR = 37$ and 257. The relation between RRR of Ag and $\kappa_{\text{Ag}}(T)$ was obtained by interpolation with reference to Cu data [6] in order to determine the RRR values of Ag particles in YBCO. Figure 4 shows the estimated RRR of doped Ag in YBCO versus Ag volume fraction f_{Ag}. RRR was almost constant for f_{Ag} below about 0.15 and then increased with increasing f_{Ag}. The value of $f_{\text{Ag}} \approx 0.15$ is consistent with the threshold volume fraction of percolation theory in a three-dimensional medium [7]. The RRR of Ag in Ag-doped Bi2212 crystals in the previous report [1] was higher than that of the present Ag-doped materials at the same Ag concentration. This probably results from the anisotropic distribution of Ag in the latter [2].

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