Superconducting Au-YBa$_2$Cu$_3$O$_7$ composites

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Superconducting Au-YBa$_2$Cu$_3$O$_7$ composites have been fabricated over a Au volume fraction range of 0–100%. Microstructure measurement indicates that Au and YBa$_2$Cu$_3$O$_7$ form well-separated phases. The superconducting transition temperature $T_c$ was found unaffected by the presence of Au. The composites exhibit low normal state resistivity and much improved ductility. The results suggest that Au is an excellent metal host for making ultrafine YBa$_2$Cu$_3$O$_7$ granular solids.

The newly discovered high $T_c$ superconducting ceramic oxide (YBa$_2$Cu$_3$O$_7$) has attracted unprecedented research activities worldwide. However, YBa$_2$Cu$_3$O$_7$ granular composite solids, in which small superconducting particles are embedded in either a metal or an insulator host, have not yet been successfully fabricated. Superconducting granular solids are a very useful medium in the studies of finite-size effects on superconductivity, thermal fluctuation effects, etc. Technologically, metallic superconductor composites offer the attractive features of a current shunt, reduced resistivity, improved mechanical properties, and environmental resistance. However, the fabrication of high $T_c$ superconducting composites faces a major obstacle. The superconducting properties depend critically on the composition, structure, and proper oxygen content. The oxides need to be processed above 900 °C in an oxygen atmosphere in order to achieve these properties. Almost all metal hosts would oxidize severely if subjected to such an adverse environment. In addition, the superconducting oxide will itself react with most metals or insulators, resulting in deteriorated superconducting properties or loss of superconductivity altogether.

In this work we will describe the fabrication of a high $T_c$ superconducting gold-YBa$_2$Cu$_3$O$_7$, composite system that can survive all of the stringent processing conditions. In this system a much lower normal-state resistivity and improved ductility have been achieved, while the high $T_c$ superconducting properties have been retained. The presence of Au increases the lattice parameters substantially, but is not seen to compromise the superconducting properties notably. The realization of this composite system opens the way to make ultrafine YBa$_2$Cu$_3$O$_7$ granules useful for fundamental studies. Our study also suggests the suitability of using Au buffer layers for YBa$_2$Cu$_3$O$_7$ thin-film devices.

Composite samples were prepared by mixing the YBa$_2$Cu$_3$O$_7$ powder with desired quantities of fine gold powder (99.9%, 325 mesh). The mixture was cold pressed into a thin disk and fired at 950 °C in flowing O$_2$ for at least 15 h before slow cooling at a rate less than 2 °C/min. The resulting disks were much less brittle than the pure superconducting material.

Figure 1 displays a few representative scanning electron micrographs; the dark regions are YBa$_2$Cu$_3$O$_7$ grains and the white regions the Au grains.

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FIG. 1. SEM micrographs of Au-YBa$_2$Cu$_3$O$_7$ composites with Au volume fractions $x_{Au} = 22.2\%$ (top), 55.0\% (middle), and 82.2\% (bottom). The dark regions are YBa$_2$Cu$_3$O$_7$ grains and the white regions the Au grains.
measurements were performed in a SQUID magnetometer. Figure 2 displays the variation of \( T_{\text{c, zero}} \) and \( T_{\text{c, SQUID}} \) with Au volume fraction \( (x_{\text{Au}}) \). The values of \( T_{\text{c, SQUID}} \) remain high (\( > 85 \) K) for Au volume fractions as high as 70%. Clearly, the presence of Au has a negligible effect on the superconducting transition temperature. The resistive transition temperature \( T_{\text{c, zero}} \) shows a different behavior. It is above 85 K for Au volume fractions up to \( x_{\text{Au}} = 40\% \) and then drops toward zero rather abruptly for \( x_{\text{Au}} \approx 70\% \), which is taken to be the critical volume fraction for connection of the superconducting material (i.e., the superconducting percolation threshold).

Because the resistivity of YBa\(_2\)Cu\(_3\)O\(_x\) is about two orders of magnitude higher than that of Au, the resistivity (\( \rho \)) in the normal state is a measure of the connectivity of the Au in the composite system. Consequently, the value of \( \rho \) will exhibit percolation behavior. Indeed, as shown in Fig. 2, the room-temperature resistivity \( \rho \) (297 K) shows a sharp drop of two orders of magnitude at the Au percolation threshold of \( x_{\text{Au}} \approx 23\% \).

The hardness of the samples was measured on a TECO M-400 Vickers Microhardness Tester. Figure 3 displays the Vickers hardness number (VHN) of these Au-123 composite materials as a function of Au content. The hardness, which is inversely related to the ductility in most materials, is seen to drop almost two orders of magnitude (from a VHN of \( \sim 475 \) to \( \sim 25 \)) starting at a Au volume fraction of \( x_{\text{Au}} \sim 20\% \).

For Au concentrations in the range of \( 20\% < x_{\text{Au}} < 60\% \) such that both the Au and YBa\(_2\)Cu\(_3\)O\(_x\) are percolating, the composite material performs as a high \( T_{\text{c}} \) superconductor while exhibiting dramatically reduced resistivity and improved ductility. The resistivity and hardness at \( x_{\text{Au}} = 40\% \) are, respectively, 0.02 and 0.3 times the values for the pure superconducting ceramic oxide; yet \( T_{\text{c, zero}} \) is 95% of the original value and \( T_{\text{c, SQUID}} \) is unchanged.

X-ray diffraction spectra were obtained from the samples with a Philips APD 3720 automated x-ray diffractometer. Each of the samples was exactly two phase: Au and orthorhombic YBa\(_2\)Cu\(_3\)O\(_x\). However, as shown in Fig. 4, there is a substantial expansion of 0.56% in the \( c \) axis in the region \( 0 < x_{\text{Au}} < 3\% \) and a slight expansion of 0.12% in the \( b \) axis. The \( a \) axis remains essentially unchanged.

There are a few possible explanations for this expansion, but some of them can be immediately eliminated. The stress-induced lattice expansion causes by the unmatched thermal expansion constants of the two materials is unlikely, because the lattice parameters of the Au remain unchanged. Moreover, the lattice expansion observed is too large to be caused by stress. Neither is the expansion caused by the loss of the oxygen content in the orthorhombic structure. Cava et al. have shown that in the YBa\(_2\)Cu\(_3\)O\(_x\) system, \( c \) expands with increasing \( \delta \). The change of \( c \) with \( \delta \) is approximately linear and can be characterized by a slope of \( (1/c)/(\Delta c/\Delta \delta) \sim -1.37\% / \text{O atom} \). In the composite system, this would correspond to \( \delta = 0.4 \). Such a level of oxygen removal is not possible if the material is to remain superconducting at temperatures above 85 K.

We are therefore led to the conclusion that the lattice-wide expansion is caused by Au entering the orthorhombic structure. As shown in Fig. 4, the \( c \)-axis expansion occurs in the region \( 0 < x_{\text{Au}} < 3\% \), beyond which it remains unchanged. This phenomenon can be best explained by the existence of a Au solubility region in the YBa\(_2\)Cu\(_3\)O\(_x\) materials, beyond which Au precipitates into separated phases. Further investigation of this anomalous lattice expansion is underway.

Most interesting of all, the superconducting transition...
temperature is not affected at all by such a large expansion of the c axis, which is intimately related to the coupling between the Cu-O$_2$ planes and the Cu-O chains. It is known that the $T_c$ of YBa$_2$Cu$_3$O$_y$ is sensitive to external pressure, with a rate of $dT_c/dP = 0.043$ K kbar$^{-1}$. The incorporation of Au, in effect, induces a uniaxial stress along the c axis. The lack of sensitivity of $T_c$ to such stress deserves further study, since it may provide valuable information as to the anisotropic electronic structures and the interlayer couplings.

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