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Magnetoresistance and percolation in Au_p–$(PrBa_2Cu_3O_7)_{1-p}$ composites

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Abstract
We examine the transverse magnetoresistance of three-dimensional bi-component composites Au_p–$(PrBa_2Cu_3O_7)_{1-p}$. This property, seldom measured on percolation systems, has been measured below 5 K and up to 1.5 T. Contrarily to well known transport properties, the magnetoresistance of these compounds exhibits a metal volume fraction's threshold p_c linked to the onset of disorder rather than to percolation.

1. Introduction

Many compounds, made of good and poor electrical conductors, are famous examples of percolation systems [1] characterized by well known properties of their electrical resistivity [2–11] or their Hall coefficient [8, 11–19], for instance. Among them, the family of Au_p–$(PBCO)_{1-p}$ composites (PBCO standing for semiconducting $PrBa_2Cu_3O_7$) has proven an excellent candidate to verify percolation theory. This has been true concerning its electrical resistivity [20, 21] (ρ) and recently measured thermoelectric power [21]. Moreover, in these cases, the large temperature variation, across the family, of the ratio of the component's electrical resistivity ρ_{Au}/ρ_{PBCO} has allowed a discussion in term of scaling, an important feature of percolation systems. Consequently, these transport properties were shown to depend on a scaling function of the form [6, 7]

$$F(x), \qquad x = r/|\Delta p|^{t+s}, \qquad \Delta p = p_M - p_c, \qquad r = \rho_M/\rho_I.$$

Here I, M, p and p_c denote respectively the bad and good conductors making the mixture, the volume fraction of M in the composite and the percolation threshold. t and s are the electrical resistivity's critical exponents.

However, few studies of the magnetoresistance (MR) of such inhomogeneous media have been conducted within the scope of percolation [22, 23]. To what extent percolation would also rule the transverse magnetoresistance of Au_p–$(PBCO)_{1-p}$ composites is the focus of this paper.

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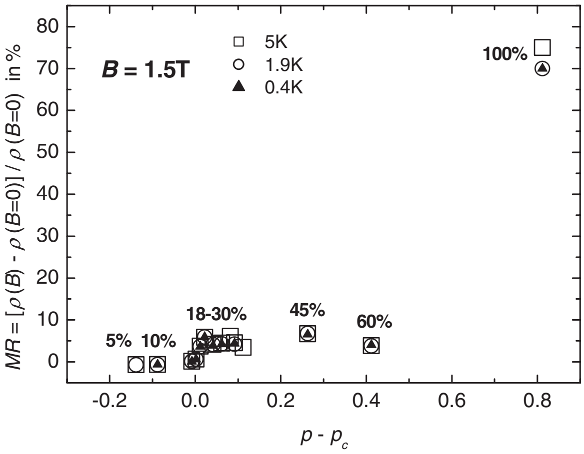

Figure 1. Transverse magnetoresistance versus $p - p_c$ of Au$_p$–(PrBa$_2$Cu$_3$O$_7$)$_{1-p}$ composites measured at 0.4, 1.9 and 5 K in a field of 1.5 T. ρ stands for the electrical resistivity, p_c is taken as 0.188 and metal volume fractions p are indicated in per cent.

2. Experimental method

The Au–PBCO compounds are bulk samples prepared into pellets by the standard solid state reaction method described previously [24]. The magnetoresistance has been measured with the standard four-probe technique using a ^{3}He cryostat (Oxford Heliox) equipped with a superconducting magnet providing a maximum field of 1.5 T. Silver paste was used to connect copper wires (cross-section: 0.5 mm) on the samples prior to a 15 min annealing step in air at 100 °C. This procedure has provided contact resistances generally between 1 and 4 Ω.

3. Results

The transverse MR, measured in the magnetic field range $B \in [-1.5\text{ T}, +1.5\text{ T}]$ and at three temperatures $T \in \{360\text{ mK}, 1.9\text{ K}, 5\text{ K}\}$, showed no hysteresis.

For the most conductive samples, at very low temperature, one reaches the limit of resolution of our instruments and the magnetoresistance then becomes naturally noisy. Still, three regions emerge (figure 1): one of high MR involving only the pure Au sample, one of intermediate MR involving samples above the electrical resistivity's percolation threshold (i.e., $p_c = 18.8\%$, [22]), and a final one, of very small MR, involving samples below that threshold.

The magnetoresistance being very sensitive to defects in general, one understands the reason behind the first two regions. A sharper threshold between the last two regions does however appear when the magnetoconductance (MC) is plotted (figure 2). One sees clearly that Au–PBCO displays a small and almost constant MC above the electrical resistivity's percolation threshold and a positive MC (negative MR) below. One could object that if one takes the occurrence of negative MR as the relevant threshold then $p = 18\%$ seems above the threshold. But one sees in figure 3 that it displays, at the lowest temperatures and for some intermediate field range, a negative magnetoresistance as well.

4. Discussion

Above p_c, the magnetoresistance displayed no clear dependence on the metal volume fraction p, contrasting with the usual power law characterizing many other properties of percolation

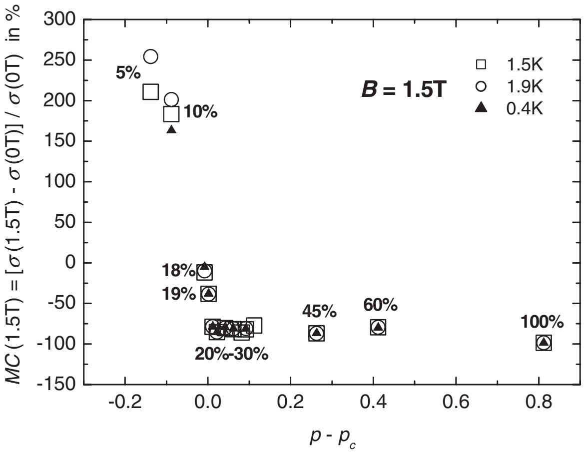

Figure 2. Transverse magnetoconductance versus $p - p_c$ of Au_p–$(PrBa_2Cu_3O_7)_{1-p}$ composites measured at 0.4, 1.9 and 5 K in a field of 1.5 T. The values of the metal volume fractions p are also indicated in per cent and σ stands for the electrical conductivity.

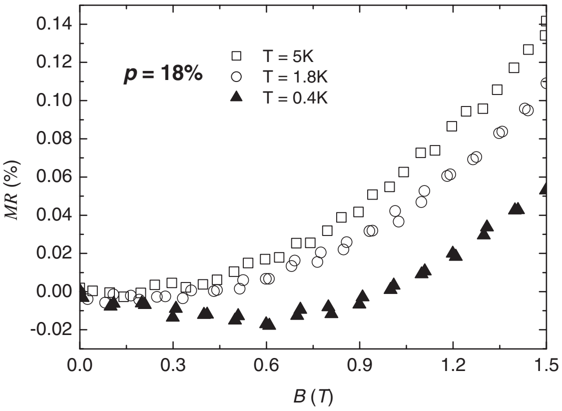

Figure 3. Field dependence of the transverse magnetoresistance of $Au_{0.18}$–$(PrBa_2Cu_3O_7)_{0.82}$ measured at 0.4, 1.9 and 5 K.

systems. Possible experimental uncertainties put aside, this seems thus more in line with the theory suggested for two-dimensional (2D) compounds [20] than with the three-dimensional (3D) theory predicting a percolation-driven magnetoresistance with explicit dependence on p and on several scaling functions [21]. However, to the authors' knowledge, no sufficiently tractable equation in the 3D case has so far been formulated, preventing thus the determination of the actual magnitude of the dependence on p.

At this point of our study we cannot offer a reasonable mechanism as to why a 2D percolation behaviour should prevail in our system. A more likely reason is that the expected 3D effect is very weak and that the observed insensitivity of the magnetoresistance with respect to p ($p > p_c$) is the result of the short-range order inherent to such composites. Except at 100% metal volume fraction, any composition above p_c is likely to present very similar mazes of twisted paths in the metallic regions. As a result, as p varies, the differences in density of these mazes are unlikely to make a clear difference in term of electron mean free path.

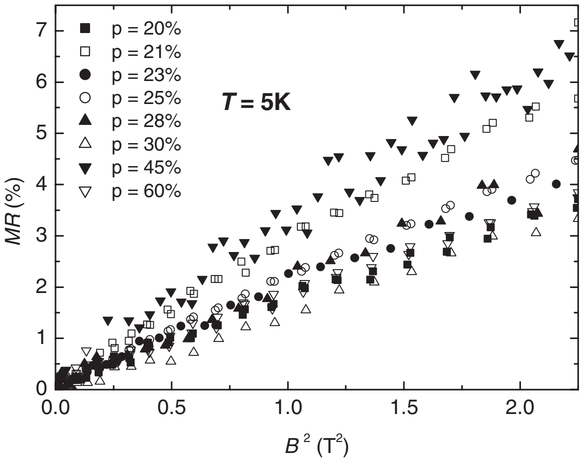

Figure 4. Transverse magnetoresistance versus B^2 of Au_p–$(\mathrm{PrBa_2Cu_3O_7})_{1-p}$ composites measured at 5 K for $p > p_c$.

One observed also no noticeable temperature dependence indicating that the metal component is likely the major contributor to the magnetoresistance above the percolation composition as the three chosen temperatures correspond to the range of temperature-independent relaxation processes (residual resistance) in metals. This seems also supported by the positive and quadratic behaviour revealed at 0.4, 1.9 and 5 K and up to 1.5 T (figure 4), a property consistent with polycrystalline noble metals.

Below p_c, negative magnetoresistance was observed, a feature also revealed in doped semiconductors and discussed as a consequence of disorder [25, 26]. Accordingly, the presence of a random potential is predicted to add a negative anomalous term to the normal positive orbital MR. The absolute value of this anomalous term is found to be proportional to B^2 ($\sqrt{B}$) for weak fields (for intermediate fields) with a magnitude decreasing with the carrier density. At higher fields, there is a field threshold, whose value decreases with increasing carrier density, and above which the normal positive MR takes over. To support that analysis, a closer look at the samples of lowest p-values reveals indeed a negative MR, first, less than quadratic in B, then, closer to a square root law with a tendency to saturate at higher field (figure 5). Finally, as p increases, one observes also a decrease of the absolute magnitude of MR up to the point that the magnetoresistance changes sign (figure 3). Although the quadratic trend does not seem to describe the measured magnetoresistance accurately at low field, disordered phenomena have been observed in the electrical resistivity of $\mathrm{PrBa_2Cu_3O_7}$, typical of variable range hopping conduction (VRH) [27], and discussed in term of localized electronic states [28–33]. To verify the possible occurrence of 3D VRH in our compounds, low-temperature measurements of the electrical resistivity of our compounds have been conducted for $p = 5$ and 10%, showing a characteristic temperature dependence (figure 6) of $\exp((T_0/T)^{-1/4})$, where T_0 is a characteristic temperature, in accordance with these disorder arguments.

5. Conclusion

Magnetoresistance measurements performed on Au_p–$(\mathrm{PrBa_2Cu_3O_7})_{1-p}$ composites have confirmed the existence of a metal volume fraction's threshold identical to those observed previously in the electrical resistivity and the thermoelectric power of this percolation system. However, no power-law dependence on p, typical of percolation, could be observed. The

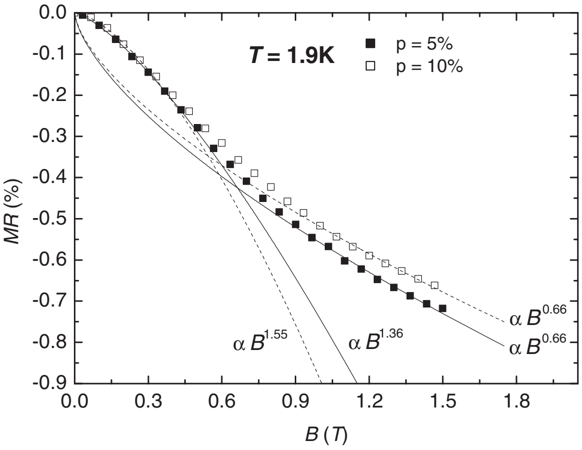

Figure 5. Field dependence of the transverse magnetoresistance of Au_p–$(PrBa_2Cu_3O_7)_{1-p}$ measured at 1.9 K for $p = 5$ and 10%. Experimental data and allometric fits (B^n) correspond respectively to scatter and line plots. The fit equations are also reported.

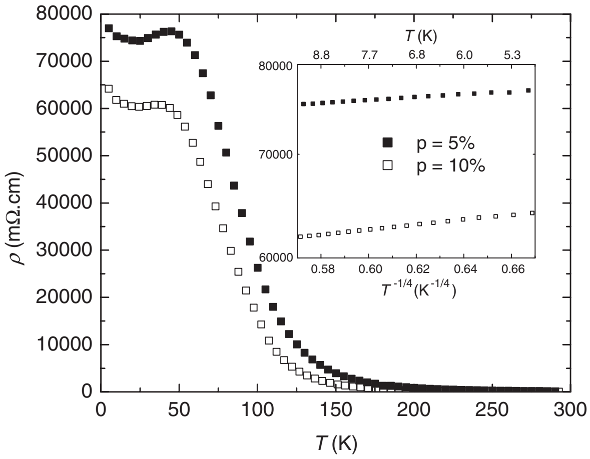

Figure 6. Resistivity versus T of Au_p–$(PrBa_2Cu_3O_7)_{1-p}$ for $p = 5$ and 10%. The inset displays a semilog plot of the low-temperature region linear in $T^{-1/4}$, as expected for 3D variable range hopping conduction.

observed threshold (separating negative from positive magnetoresistance) is suggested to relate rather to the disorder properties intrinsic to $PrBa_2Cu_3O_7$.

Acknowledgments

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