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Thermopower of non-superconducting and superconducing $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$ samples

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Abstract. The thermopower of single-phase samples of $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$ was measured from 250 K down to 10 K. The as-prepared sample was not superconducting. It had a negative thermopower at 250 K, whose magnitude increased as temperature was decreased to 95 K. A further reduction in temperature caused a decrease in magnitude of thermopower. The sign of the thermopower changes to positive at 12 K. The superconducting sample also showed the same behaviour but the change of sign now occurred at 40 K. Below 40 K, the thermopower showed a positive peak and reduced to zero at the superconducting transition. These results were compared with previous studies.

Keywords. Electron superconductor; thermopower; phonon drag; electron diffusion.

1. Introduction

$Nd_{2-x}Ce_xCuO_{4-y}$ is an electron superconductor for Ce concentrations between 0·14 and 0·18. It will be interesting to measure the thermopower of these materials and to check if the sign of the thermopower agrees with the sign of the carriers. With this aim in view, we prepared single-phase samples of $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$ and measured the thermopower of these samples in the temperature range 250–10 K.

2. Experimental

The preparation of these materials was described elsewhere (Vijayashree *et al* 1990a). The as-prepared samples were not superconducting. Samples were then heated to 850°C in a vacuum of 10^{-5} torr for 6 h. The samples so annealed were found to exhibit a superconducting transition with T_c (onset) at 24·5 K and T_c(zero) at 10·25 K. Figure 1 shows the temperature variation of resistivity of the non-superconducting and superconducting samples. Superconducting onset temperatures in these samples were also checked by magnetic susceptibility measurements in a SQUID magnetometer (figure 2).

The thermopower was measured on pellets of these samples using a set-up described earlier (Vasudeva Rao *et al* 1984).

3. Results and discussions

Figure 3 shows the temperature variation of absolute thermopower of the superconducting and non-superconducting samples of $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$. Two different samples prepared under identical annealing conditions gave similar results. From the figure , it is seen that the non-superconducting sample has a negative sign for the thermopower at 250 K. As the temperature is reduced, the magnitude of the

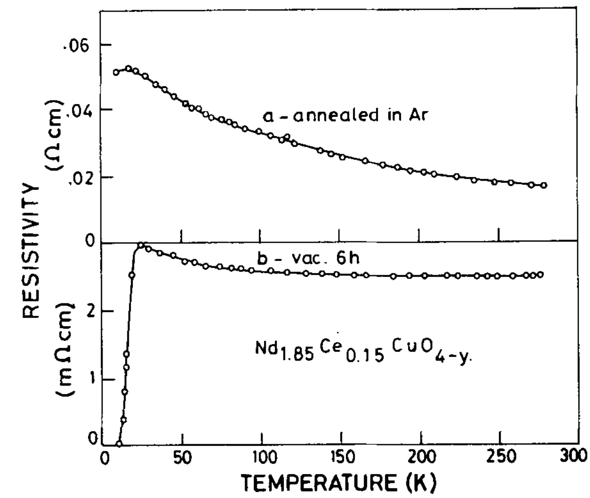

Figure 1. Resistivity vs temperature of $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$.

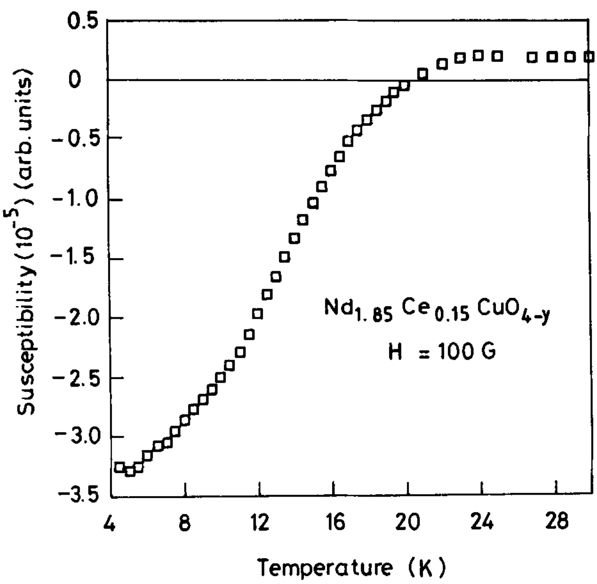

Figure 2. Susceptibility vs temperature of $Nd_{1\cdot 85}Ce_{0\cdot 15}CuO_{4-y}$.

thermopower attains a peak at around 95 K. With further reduction in temperature, the magnitude of the thermopower decreases and the thermopower undergoes a change of sign at 12 K. The same features are exhibited by the superconducting sample, but for the following differences: (a) the thermopower at 250 K of the superconducting sample is nearly five times smaller in magnitude than the thermopower of the non-superconducting sample, (b) the negative peak in thermopower of the superconducting sample occurs at around 105 K, and (c) the thermopower sign changes from negative to positive at 40 K in the superconducting sample.

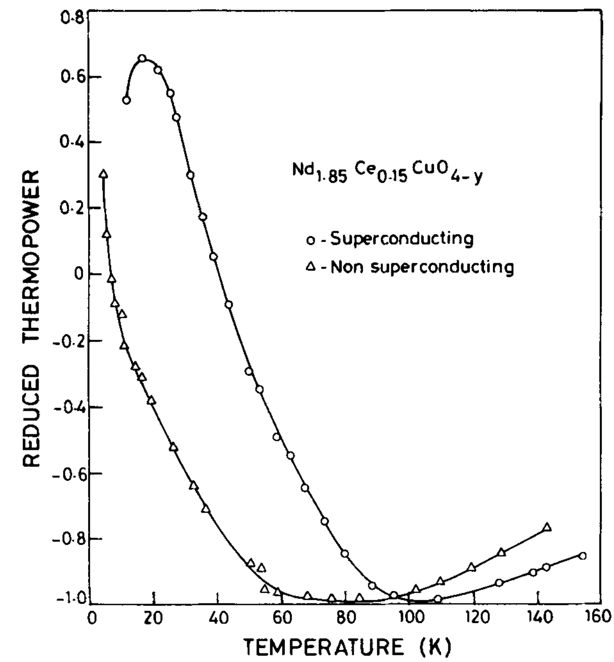

Figure 3. Thermopower vs temperature of $Nd_{1\cdot85}Ce_{0\cdot15}CuO_{4-y}$.

Lim *et al* (1989) measured the thermopower of $Nd_{1\cdot85}Ce_{0\cdot15}CuO_{4-y}$ and $Pr_{1\cdot85}Ce_{0\cdot15}CuO_{4-y}$. These samples were sintered at 1150°C for 12 h and annealed at 1000°C in Ar atmosphere for 10 h. For the Nd sample studied by them, the resistivity dropped at 24 K, but did not go to zero down to 4·2 K, probably due to incomplete oxygen deficiency. They found the thermopower to be positive and small. The thermopower increased when the temperature was increased from 10 K, reached a maximum value of 0·7 μV/K around 50 K, and then decreased and reached a plateau in the temperature region 100–250 K. With further rise in temperature, thermopower decreased sharply.

On the other hand, Lopez-Morales and Grant (1989) reported that the thermopower of the parent compound Nd_2CuO_{4-y} was negative at room temperature and underwent a change of sign at around 70 K. But in $Nd_{1\cdot85}Ce_{0\cdot15}CuO_{4-y}$, these authors found the thermopower to be negative and constant with a value of around 1·8 μV/K, which decreased smoothly to zero at the superconducting transition. They did not observe a change in sign of thermopower at low temperatures. These authors conclude that the positive thermopower arises from a non-uniform distribution of Ce.

Takagi *et al* (1989) found the thermopower to be around -2μV/K and temperature-independent down to the onset of superconducting transition. The results of Lim *et al* (1989) contrast sharply with those of Lopez-Morales and Grant (1989) and Takagi *et al* (1989). Our results do not agree with either of them.

Sony corporation workers (1989) reported that the sign of thermopower was negative at room temperature and became positive just above the superconducting transition. These results are in agreement with ours.

Our results can be interpreted as arising from a combination of diffusion and phonon drag contributions. The phonon drag contribution appears to be positive in these samples. The electron diffusion term in the non-superconducting sample appears to be very large and compensates for the phonon drag term at a low temperature (around 12 K). The superconducting sample has a considerably smaller electron diffusion term. The removal of oxygen is not expected to alter the phonon drag contribution drastically. Hence, the electron diffusion term compensates the phonon drag term at a much higher temperature (around 40 K).

Another possible explanation could be that we have both electrons and holes contributing to the thermopower as is believed to happen in the Tl and Bi compounds (Vijayashree *et al* 1990b). In such a case, the sign of thermopower is not an unambiguous indication of the sign of the charge carrier responsible for superconductivity. Measurements on samples with other Ce concentrations showed similar results (Vijayashree *et al* 1990c).

Lopez-Morales and Grant (1989) performed an EPMA of their sample and concluded that the Ce concentration in the sample varied from point to point giving rise to inhomogeneity in the sample. They also believed that the region of inhomogeneity was Nd_2CuO_{4-y} with low amounts of Ce. One may attribute the changeover from negative to positive thermopower to the presence of such low concentrations of Ce. However, it is not clear as to how the thermopower of such an inhomogeneous phase will behave.

4. Conclusions

The thermopower of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ has been measured both for the non-superconducting and superconducting samples. In both cases, the thermopower changes from negative to positive as the temperature is reduced. The cross-over temperature for the non-superconducting sample is lower than that for the superconducting sample. Our results are in total disagreement with those of Lim *et al* (1989), whose findings showed positive thermopower at all temperatures down to the superconducting transition. It is in partial agreement with the results of Lopez-Morales and Grant (1989) in that, the sign, magnitude and trend of temperature variation of thermopower are similar down to about 50 K for the superconducting sample. However, we observe a definite change of sign in thermopower at 40 K which is not seen by Lopez-Morales and Grant (1989). Our results can be understood in terms of a positive phonon drag contribution and a negative electron diffusion contribution.

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