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## The Hall effect of the superconducting oxides (Bi, Pb)<sub>2</sub>Ca<sub>2</sub>Sr<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub>

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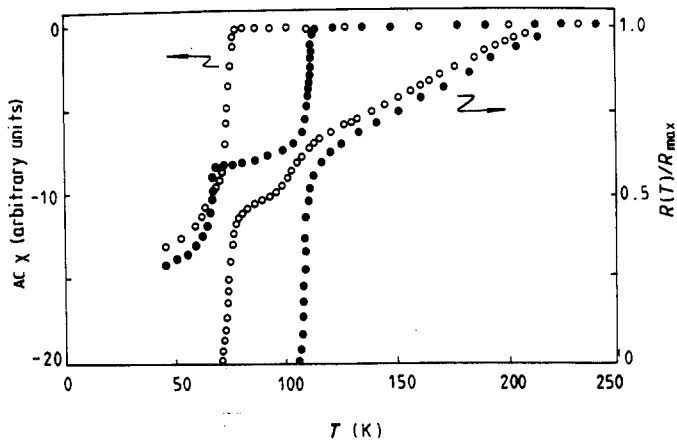
**Abstract.** The temperature dependence of the Hall coefficient  $R_H$  has been measured between 50 and 250 K for the superconducting compounds with nominal compositions  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$  and  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ . It is found that the Hall number,  $n_H$ , is linearly  $T$ -dependent and field-independent in the normal state and decreases rapidly on doping with lead. This result is quite in agreement with the recent theoretical calculation proposed by Xing and co-workers. A large anomaly in  $R_H$  is observed around  $T_c$ , which is discussed in terms of the intergranular structure of this material.

### 1. Introduction

The Hall effect in both ceramic and single-crystal superconducting  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-y}$  compounds has been investigated by many groups. The data show strong anisotropies and unusual temperature dependence of electronic properties. These may be helpful in understanding the novel mechanism for high- $T_c$  superconductivity. An important question is whether or not the extraordinary properties are common characteristics for all oxide cuprates, which have different crystalline and electronic structures. In particular, the newly discovered Bi-site compounds without any rare-earth elements exhibit many different features compared to those of previously known 90 K superconductors; there are neither Cu–O chains nor Cu–O bonds linking the Cu–O planes in them. Does this act on their transport properties in the normal state? In this paper, we make a contribution towards answering this question through a study of Hall effect in the Pb-doped BiCaSrCuO system.

### 2. Experimental details

Samples with nominal compositions  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$  and  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$  were selected for the study of the Hall effect. Their synthesis, phase and crystalline structure features were presented and discussed by Chen *et al* (1988). Their basic superconducting properties are reproduced in figure 1. The zero resistance temperature  $T_{ce}$  of the unleaded sample is 75.5 K. A small drop in resistance at about 110 K is observed; this



**Figure 1.** Temperature dependence of the resistivity and AC magnetic susceptibility for  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$  (○) and  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$  (●).

means the sample includes a little of the higher- $T_c$  superconducting phase in addition to the main 80 K phase. The  $T_{ce}$  of the Pb-doped sample is as high as 107 K, and the measurement of the AC susceptibility indicates that the amount of the higher- $T_c$  (107 K) superconducting phase is much larger than that of the lower  $T_c$  (67 K) one.

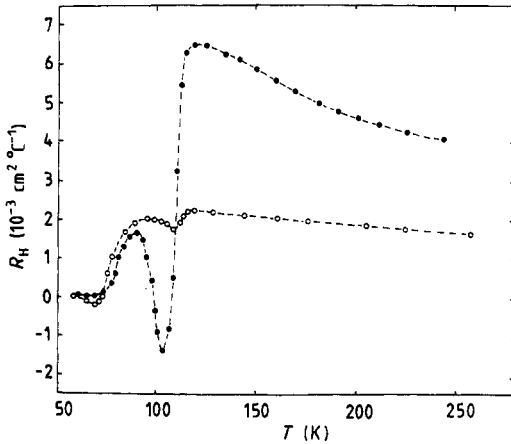
The samples were cut into bars of  $12 \times 4 \times 0.7 \text{ mm}^3$ ; four silver electrodes were evaporated in a standard Hall configuration and copper leads were soft-soldered to the electrodes. This procedure ensures low contact resistances of less than  $10^{-4} \Omega \text{ cm}^{-2}$ . The temperature dependences of the Hall voltage ( $V_H$ ) were measured in a magnetic field of 3 T, and the field dependences of  $V_H$  were obtained in the fields of from 1 to 5 T at 135 K only. To avoid errors due to the non-symmetry of the Hall potential probes, the Hall coefficient was calculated for both magnetic-field and measuring-current directions. The sample temperature was controlled to within 2 mK during the collection of the Hall voltage data. The measuring current density is about  $2 \text{ A cm}^{-2}$ , and the voltage resolution is better than  $10^{-8} \text{ V}$ .

### 3. Results and discussion

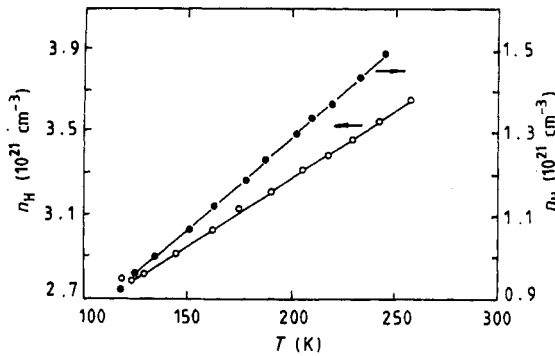
#### 3.1. The anomaly in $R_H(T)$ and its explanation

The Hall coefficient  $R_H$  against temperature curves for both samples probably do have some common characteristics, as shown in figure 2.  $R_H$  is positive in the normal state, and increases slowly as the temperature drops down to the  $T_c$  onset, then, via an anomalous region, decreases to zero, as expected for a superconductor. To a certain extent, this behaviour is similar to that in ceramics of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  and  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ , as reported by Zhao *et al* (1987) and Hundley and Zettl (1987), respectively. Therefore, it would seem that the anomaly of  $R_H(T)$  around  $T_c$  is a common feature in the high  $T_c$  superconducting ceramics. We propose that it is associated with the complex microstructure of the ceramic materials, that is, that the valleys of  $R_H(T)$  below  $T_c$  originate from grain boundaries.

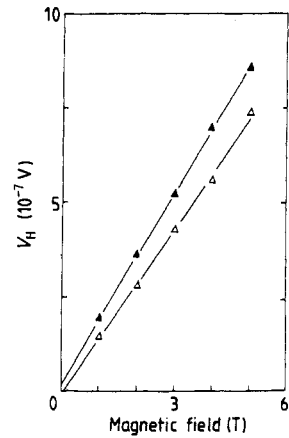
If such an anomalous phenomenon is really related to the grain boundaries, it should not occur in a single crystal. Recent work on both ceramic and single crystal



**Figure 2.** Temperature dependence of the Hall coefficient for both samples:  $\circ$ ,  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ ; and  $\bullet$ ,  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ .



**Figure 3.** The variation of Hall number against temperature:  $\circ$ ,  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ ; and  $\bullet$ ,  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ .



**Figure 4.** The dependence of Hall voltage on the applied magnetic field at 135 K;  $\triangle$ ,  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ ; and  $\blacktriangle$ ,  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ .

$\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  compounds reported by Thier and Winzer (1988) has strongly supported our proposition. No valley on  $R_H(T)$  below  $T_c$  was observed on the single crystal, but there is a large one on the bulk samples.

### 3.2. The Hall number, $n_H$

The Hall number  $n_H = 1/eR_H$  is linearly temperature dependent in the normal state (as shown in figure 3). The formula  $n_H(T) = n_0(1 + aT)$  is well satisfied for both samples, where  $n_0$  and  $a$  are  $0.38 \times 10^{21} \text{ cm}^{-3}$  and  $0.012 \text{ K}^{-1}$  respectively for the sample of  $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ ; and  $1.95 \times 10^{21} \text{ cm}^{-3}$  and  $0.0034 \text{ K}^{-1}$  for  $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_y$ . The  $n_H$  at 250 K is  $1.49 \times 10^{21} \text{ cm}^{-3}$  for Pb-doped sample and  $3.66 \times 10^{21} \text{ cm}^{-3}$  for the other one. Obviously, the carrier concentration is reduced by doping with lead.

The experimental results, e.g.  $T$ -dependence of the Hall number, are in good agreement with the two-band model with an extra carrier source proposed by Xing and Ting (1988) recently. In this model, the Hall number can be described by the equations

$$n_H(T) = n_0(0)(1 + T/T^*) \quad (1)$$

$$T^* = \pi d N_1 / (m_e + m_h) \quad (2)$$

where  $N_1$  represents the density of the localised states provided by oxygen vacancies,  $d$  the lattice parameter along the  $c$ -axis, and spacing  $m_e$  and  $m_h$  the effective mass of electron and hole, respectively. From these formulae, and assuming  $m_e$  and  $m_h$  to be identical for both samples, it is quite reasonable to suppose that the Hall number declines with doping lead due to the partial substitution of  $\text{Pb}^{4+}$  for  $\text{Bi}^{3+}$  leading to a decrease of vacancies in this oxygen-deficient material. In addition, we deduce that  $dn_H(T)/dT$  is proportional to  $1/N_1$  or  $a$ , which agrees qualitatively with the experimental result.

It must be pointed out that the linear  $T$ -dependence, and even the magnitude and the sign of Hall number  $n_H$  coincide with that of  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-y}$  compounds reported by Zhang *et al* (1987) and Tanaka *et al* (1987). This coincidence may imply that there are similar band structures that originate from the conductive tunnels of these materials. Indeed, the electronic structure proposed by Massidda *et al* (1988) for  $\text{Bi}_2\text{CaSr}_2\text{Cu}_2\text{O}_8$  indicates that the band structure and  $N(E_F)$ , the density of states at  $E_F$ , even for the highly 2D-Fermi surface that is closely related to the transport process as well as to the superconductive mechanism, do not essentially change compared with the  $\text{ReBa}_2\text{Cu}_3\text{O}_{7-y}$  compounds except for the Bi–O planes, which play a role similar to that of the Cu–O chains. On the other hand, a weak  $T$ -dependence on Hall coefficient was observed by Hundley and Zettl (1987) in  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ . The  $T_{ce}$  of this material is only 36 K, but the Hall number  $n_H$  at room temperature reaches  $6 \times 10^{21} \text{ cm}^{-3}$ . This is much larger than that of the other two kinds of high- $T_c$  oxide superconductors. Perhaps, this means that the electron–phonon interaction in the latter is much stronger than that in  $\text{K}_2\text{NiF}_4$  type compounds, and implies that some novel scattering mechanisms exist in the present materials.

### 3.3. Relationship between the Hall voltage and the applied magnetic field

The Hall voltage against the applied magnetic fields curves for both samples at 135 K are reproduced in figure 4. It can be seen that the  $V_H$  is proportional to the magnetic field. This means that the Hall coefficient or the carrier concentration in a magnetic field of up to 5 T is field-independent in the normal state. It is possible that this observation may rule out certain asymmetric magnetic scattering mechanisms.

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