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LETTER TO THE EDITOR

Two lower critical fields $H_{c_1}(0)$ in the high-T_c superconducting $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ system

Liu Hongbao, Mao Xianglei, Zhang Weijie, Mao Zhiqiang, Li Bi-you, Chen Zhaojia and Zhang Yuheng
Department of Physics, University of Science and Technology of China, Hefei, Anhui, People's Republic of China

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Abstract. Magnetisation curves for the $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ system at different temperatures are reported. There are two phases with different critical temperatures. Corresponding to the two phases, two lower critical fields $H_{c_1}(0)$ are obtained.

After a new type of superconducting Bi–Sr–Cu oxide system had been found by Michel and co-workers [1], Chu and co-workers [2] observed superconductivity up to 120 K in multi-phase Bi–Al–Ca–Sr–Cu–O and Bi–Sr–Ca–Cu–O systems. However, it is difficult to obtain a superconductor with a zero-resistance transition temperature higher than 90 K in this system. Recently Green and co-workers [3] have succeeded in obtaining a superconducting system with a zero-resistance transition temperature higher than 100 K by chemical substitution of Pb in the Bi–Sr–Ca–Cu–O system. In this system, Pb replaces Bi in the unit cell, the superconducting phase having the same structure as the alloy without lead [4]. It has been found that the controlled addition of Pb to the alloy leads to the elimination of the resistance step, although the step in the diamagnetic susceptibility remains [3]. In this Letter we report some results concerning superconductivity and magnetisation for the lead-substituted samples. The $H_{c_1}(T)$ curve and two critical fields $H_{c_1}(0)$ have been obtained.

The samples were prepared by solid-state reaction of Bi_2O_3, CuO, $SrCO_3$, $CaCO_3$ and PbO in air. The compositions were $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ with (A) $x = 0.2$; (B) $x = 0.4$ and (C) $x = 0.6$. The mixture was heated at 830 °C for 15 h. After being ground and pelletised, it was sintered at 870 °C for 68 h, and this treatment was followed by furnace cooling. A four-lead method was employed for the resistance measurements. Magnetisation curves were measured by the technique described in [5]. An x-ray diffractometer was used, with Cu Kα radiation.

The temperature dependence of R for these samples is shown in figure 1. The superconducting transition temperatures of the compounds $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ for $x = 0.2$, 0.4 and 0.6 were all found to be 107 K. Figure 2 shows the powder x-ray diffraction experimental results for samples A, B and C. All the x-ray diffraction curves contain two phases, I and II; they have exactly the same structure as the 85 K and 110 K phases in the Bi–Sr–Ca–Cu–O system. Part of the low-field diamagnetisation of sample C (M) is shown in figure 3(a). A peak appears in each curve, indicating that the sample

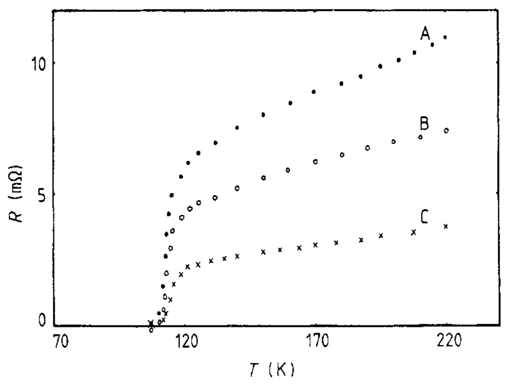

Figure 1. Resistivity against temperature for $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$, with x = (A) 0.2, (B) 0.4, (C) 0.6.

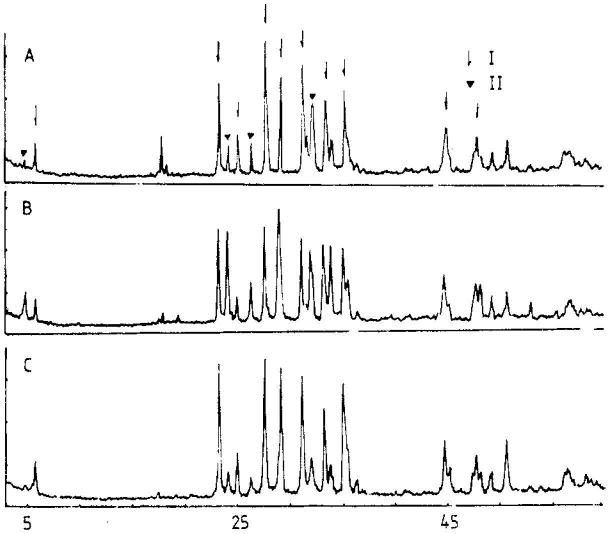

Figure 2. Powder x-ray diffraction curves for samples A, B and C.

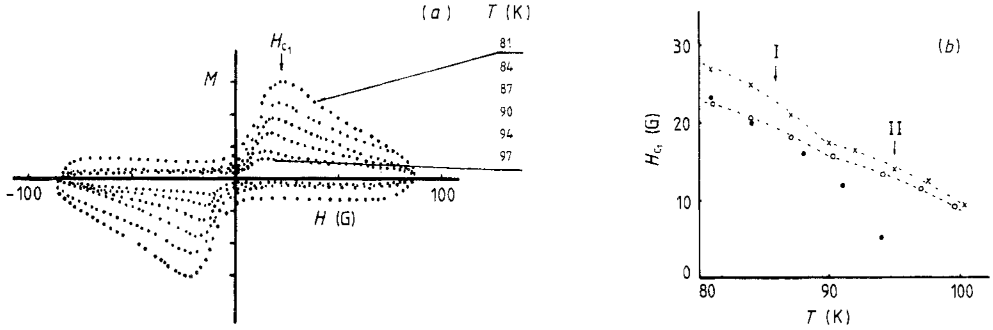

Figure 3. (*a*) Diamagnetisation curves, M–H, for sample C; (*b*) $H_{c_1}(T)$ for samples A (●), B (×) and C (○).

is indeed a type-II superconductor. The peaks were identified as the lower critical fields, $H_{c_1}(T)$ ($H_{c_1}(T)$ is often defined as the field for which M is no longer linear in H). In figure 3(*b*) the temperature dependence of H_{c_1} is shown. For samples B and C the curves for $H_{c_1}(T)$ were each obtained by combining two parts.

All of the zero-resistance temperatures were close to 107 K. The powder x-ray diffraction experiments show that the samples contain two phases, with $c = 30$ Å and $c = 38$ Å. It is suggested that the phase with $c = 38$ Å corresponds to the high-critical-temperature phase [3]. In the Pb-doped sample, the increase in c from 30 Å to 38 Å is due to the insertion of two Cu–O and two Ca layers inside the inter-growth structure [6–8]. It is also thought that the substitution of Pb in the Bi–Sr–Ca–Cu–O system raised the zero-resistance transition temperature, perhaps by increasing the Cu^{3+}/Cu^{2+} ratio or stabilising the 110 K phase [9–11].

It is clear that the $H_{c_1}(T)$ curve is composed of two parts for sample B and sample C, but one part for sample A. The relation

$$H_{c_1}(T) = H_{c_1}(0)(1 - T^2/T_c^2) \qquad (1)$$

was used to fit the experimental data. It can be obtained by using the two values of T_c and $H_{c_1}(0)$ for each sample. For sample B, $T_{cL} = 102$ K and $H_{c_1L}(0) = 78$ G; $T_{ch} = 117$ K and $H_{c_1h}(0) = 44$ G. For sample C, $T_{cL} = 104$ G and $H_{c_1L}(0) = 54$ G; $T_{ch} = 117$ K and $H_{c_1h}(0) = 38$ G. This means that samples B and C contain two phases; the critical temperature of the low-T_c phase is 100 K and that of the high-T_c phase is about 120 K. The lower critical field $H_{c_1}(0)$ of the high-T_c phase is lower than that of the low-T_c phase. It is known that in the Bi–Sr–Ca–Cu–O system there are two phases, with $T_c = 85$ K and $T_c = 110$ K. Because of the substitution of Pb in the Bi–Sr–Ca–Cu–O system, the zero-resistance temperature rises to 107 K. Our results for the two superconducting phases (100 K and 120 K) lead us to suggest that, because of the substitution of Pb in the Bi–Sr–Ca–Cu–O system, the 80 K phase becomes the 100 K phase and the 100 K phase becomes the 120 K phase, although the structure is not changed. So, replacing Bi by Pb not only stabilises the structure with $c = 38$ Å, but also changes the electronic state of the Bi–Sr–Ca–Cu–O system. These two factors raise the value of T_c for the 80 K phase and the 110 K phase in the Bi–Sr–Ca–Cu–O system. The critical temperature of a single phase with $c = 30$ Å would be 100 K and with $c = 38$ Å would be 120 K in the $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ system. Samples A, B and C are all mixtures of two phases, so the critical temperature in the $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ system is lower than 120 K.

In summary, in the $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$ system there are two phases with critical temperatures of 100 and 120 K. Doping the Bi–Sr–Ca–Cu–O system with Pb not only stabilises the structure with $c = 38$ Å, but also increases the superconductivity of the 85 and 110 K phases in the Bi–Sr–Ca–Cu–O system. In this system, two lower critical fields $H_{c_1}(0)$ were obtained.

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