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Physica B 374–375 (2006) 75–78
PHYSICA B
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Magnetic and superconducting phase diagram in oxybromite cuprate $Ca_{2-x}Na_xCuO_2Br_2$

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Abstract

A comprehensive magnetic and superconducting phase diagram determined by muon spin rotation/relaxation (μSR) is presented for $Ca_{2-x}Na_xCuO_2Br_2$ which has apical bromine atoms. Evidence for antiferromagnetic (AF) order in lightly doped samples and that for quasi-static spin glass (SG)-like state in moderately doped ones are obtained by ZF-μSR at low temperatures. While the phase diagram is qualitatively similar to that in typical 2-1-4 cuprates including $Ca_{2-x}Na_xCuO_2Cl_2$ and $La_{2-x}Sr_xCuO_4$, it exhibits a slight shift (expansion) over the x axis.

Keywords: $Ca_{2-x}Na_xCuO_2Br_2$; Halogen atom; Magnetic phase diagram; μSR

1. Introduction

Since the discovery of copper oxide superconductors, numerous experiments and theoretical studies have been performed to elucidate the role of apical atoms over the CuO_2 planes. Al-Mamouri et al. [1] reported the superconductivity in $Sr_2CuO_2F_{2+\delta}$ at $T_c = 46$ K by substituting apical O^{2-} with F^-. Subsequently, Hiroi et al. [2] demonstrated superconductivity in $Ca_{2-x}Na_xCuO_2Cl_2$ (Na–CCOC) with $T_c = 29$ K over an optimally hole-doped region ($0.15 < x < 0.20$). Recently, Zenitani et al. [3] succeeded in mapping out a comprehensive phase diagram of superconductivity in $Ca_{2-x}Na_xCuO_2Cl_{2-y}Br_y$, focusing on the relationship between the superconductivity and Cu and Cl/Br distance. The crystal structure of this compound is similar to that of La_2CuO_4, but all the apical oxygen atoms are replaced by Cl/Br atoms.

In the process of this study, a peculiar feature has been observed in $Ca_{2-x}Na_xCuO_2Br_2$ (Na–CCOB) that definitive superconductivity with $T_c = 19$ K develops over an optimally hole-doped region $x \sim 0.25$ which is substantially shifted from that of typical copper-oxide systems. Unfortunately, it is difficult to determine the magnetic critical temperature (T_N) for the lightly hole doped Na–CCOB (including $x = 0$, CCOB) by the conventional magnetic susceptibility measurements because of extremely weak magnetic response due to the non-distorted CuO_2 plane. In order to elucidate the magnetic properties in the low-doping region, we have conducted μSR experiment to map out the comprehensive magnetic phase diagram of Na–C-COB.

In this paper, we report on the magnetic and superconducting phase diagrams of Na–CCOB determined, respectively, by μSR and magnetic susceptibility measurements. The spontaneous precession signal in the ZF-μSR time spectra expected for the occurrence of AF long range order has been observed in the undoped and slightly Na-doped samples ($0 < x < 0.03$) at low temperatures. While no precession signal was identified in those with $0.04 < x < 0.15$, evidence was found for a quasi-static spin

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doi:10.1016/j.physb.2005.11.019

glass (SG)-like state at lower temperatures. Moreover, development of superconducting state was observed in the sample with $x \geqslant 0.15$. These characteristic doping levels are higher than those found in typical 2-1-4 cuprates, suggesting a shift (or expansion) of the entire phase diagram along the x axis.

2. Experimental results

A polycrystalline sample of CCOB was prepared by the solid state reaction under Ar atmosphere with stoichiometric mixture of $CaBr_2$(99.9%) and CuO(99.9%) as a starting material. Those of Na–CCOB were synthesized under an ultra-high pressure (~5.5 GPa). The samples were characterized by means of magnetization and powder X-ray diffraction measurements. We have confirmed that the no impurity phase was detected for any composition in the Na doping range and that the lattice parameter systematically decreased with increasing x. Unfortunately, the actual carrier concentration is yet to be determined quantitatively at this stage, and therefore the sodium concentration x in this paper refers only to a nominal composition.

μSR measurements were performed at the Tri-University Meson Facility (TRIUMF, Canada), Meson Science Laboratory (KEK-MSL, Japan) and RIKEN-RAL Muon Facility (RIKEN-RAL, UK). ZF- and LF-μSR spectra were obtained at temperatures between 2 K and ambient temperature. TF-μSR measurements have been employed at ambient temperature for the correction of instrumental asymmetry. The superconducting phase was determined by a superconducting quantum interference device (SQUID) magnetometer.

Fig. 1 shows the ZF-μSR time spectra in the parent material CCOB at several temperatures. The time spectra at low temperatures exhibit spontaneous oscillation expected for the occurrence of AF long range order. However, compared with the cases of other halogen cuprates (CCOC [4] and $Sr_2CuO_2Cl_2$(SCOC) [5]), it is notable that the precession signal is relatively weak probably due to smaller volume fraction of AF state. These spectra are well reproduced by the following form,

$$P_z(t) = \sum_{i=1}^{n} A_i \exp[-(\sigma_i t)^2] \cos(2\pi f_i t + \phi) + A_n \exp[-(\sigma_n t)^\beta], \quad (1)$$

where A_i and A_n refer to the asymmetry for the oscillating and non-oscillating components. $\sigma_{i,n}$ is the relaxation rate, f_i is the muon spin precession frequency, ϕ is the initial phase and β is the power of the exponent.

The temperature dependence of high (f_1) and low (f_2) frequency components is shown in Fig. 2. The f_1 exhibits a steep enhancement with decreasing temperature below 200 K, while its oscillating amplitude disappears above ~200 K. Moreover, it turned out that a precession signal obtained by fitting analysis exhibits further splitting into two components below 180 K. It is interesting to note that one of those components (f_2) takes a value common to that in CCOC and LSCO [6], and f_1 is close to that in CCOC and SCOC. Solid curve for f_1 in Fig. 1 is the fitting result with a form, $A(T_N - T)^\gamma$, which yields $T_N = 186.5(5.0)$ K. The obtained T_N is considerably smaller than that in both CCOC and SCOC which exhibit T_N~260 K as determined by μSR measurements [4,5]. The small T_N in CCOB implies that the interplanar exchange interaction in CCOB is suppressed in comparison with that in CCOC and SCOC due to the increase of c axis (~2 Å) by substituting an apical Cl^- with Br^-. Moreover, ZF-μSR data for the lightly Na doped compounds ($x = 0.02, 0.03$) show clear oscillating signal at low temperatures. A fitting procedure similar to that for CCOB was employed to the data analysis for these samples. As a result, we found a steep decrease of T_N with increasing sodium concentration; $T_N = 168.5(7.0)$ K for $x = 0.02$ and 117.1(8.0) K for $x = 0.03$.

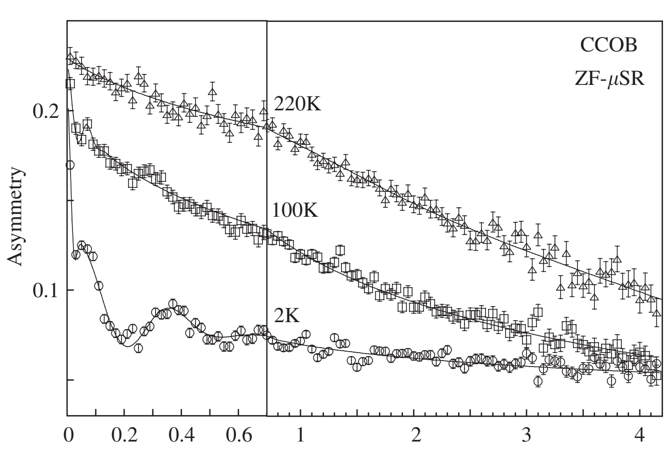

Fig. 1. ZF-μSR time spectra in parent material CCOB at several temperatures. Solid curves are results of fitting by Eq. (1).

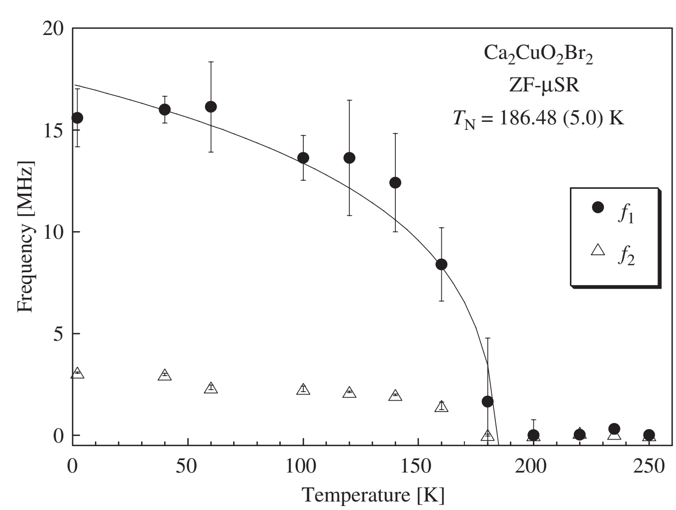

Fig. 2. Temperature dependence of muon spin precession frequency f_1 (solid symbol) and f_2 (open symbol) in Eq. (1). Solid curve is the result of fitting by $A(T_N - T)^\gamma$.