

Abstract Submitted
for the MAR08 Meeting of
The American Physical Society

Precise control of the Na nonstoichiometry in Na_xCoO_2 YOSHIHIKO OKAMOTO, ATSUSHI NISHIO, YOKO KIUCHI, ZENJI HIROI, Institute for Solid State Physics, Univ. of Tokyo — Na_xCoO_2 has been attracting great interests in terms of a correlation between its electronic properties and Na nonstoichiometry. Many groups have reported that a Na rich phase ($x \sim 0.7$) is a Curie-Weiss metal and a poor phase ($x \sim 0.3$) is a Pauli paramagnetic metal. The origin of this difference, however, has not been confirmed yet, mainly because of difficulty in controlling precisely the Na nonstoichiometry. We succeeded in synthesizing a series of polycrystalline samples of Na_xCoO_2 with well-controlled Na content by the solid-state reaction instead of the solution reaction previously used. We prepared polycrystalline Na_xCoO_2 ($0.58 \leq x \leq 0.63$) by a solid-state reaction of $\text{Na}_{0.71}\text{CoO}_2$ and $\text{Na}_{0.5}\text{CoO}_2$ at 200°C . Furthermore, fine tuning of Na content in the range of $0.62 < x < 0.63$ was carried out by a solid-state reaction of $\text{Na}_{0.62}\text{CoO}_2$ and $\text{Na}_{0.63}\text{CoO}_2$. Magnetic susceptibility of Na_xCoO_2 exhibited Curie-Weiss behavior at $x \geq 0.621$ while nearly temperature independent paramagnetism at $x \leq 0.620$. Such a drastic change of magnetism clearly indicates that the magnetic phase boundary is located in an extremely narrow range around $x = 0.62$. We think that the difference originates from a change of the Fermi surface topology with electron filling probably associated with the dip in the a_{1g} band near the Γ point.

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Date submitted: 12 Dec 2007

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