Angular-dependent Magnetoresistance Oscillations in Na$_{0.48}$CoO$_2$ Single Crystal

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We report measurements of the c-axis angular-dependent magnetoresistance (AMR) for a Na$_{0.48}$CoO$_2$ single crystal, with a magnetic field of 10 T rotating within Co-O planes. Below the metal-insulator transition temperature induced by the charge ordering, the oscillation of the AMR is dominated by a two-fold rotational symmetry. The amplitudes of the oscillation corresponding to the four- and six-fold rotational symmetries are distinctive in low temperatures, but they merge into the background simultaneously at about 25 K. The six-fold oscillation originates naturally from the lattice symmetry. The observation of the four-fold rotational symmetry is consistent with the picture proposed by Choy et al. that the Co lattice in the charge ordered state will split into two orthonhombic sublattices with one occupied by Co$^{3+}$ ions and the other by Co$^{4+}$ ions. We have also measured the c-axis AMR for Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$ single crystals, and found no evidence for the existence of two- and four-fold symmetries.

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Recently great attention has been paid to the investigation of physical properties of the newly discovered superconductor Na$_{0.35}$CoO$_2$·1.3H$_2$O with $T_c \sim 4.5$ K. Until now it is the only known layered transition metal oxide which exhibits superconductivity other than high-temperature cuprates and Sr$_{2}$RuO$_{4}$. The non-hydrated host oxide Na$_x$CoO$_2$ is known for its large thermoelctric power. It is a two-dimensional cobalt oxide. Co atoms form a triangular lattice rather than a square one as in high-$T_c$ cuprates and Sr$_{2}$RuO$_{4}$. The charge ordering is suppressed along the Co-O planes, the charge ordering is suppressed into the background simultaneously at about 25 K. The six-fold oscillation originates naturally from the lattice symmetry. The observation of the four-fold rotational symmetry is consistent with the picture proposed by Choy et al. that the Co lattice in the charge ordered state will split into two orthonhombic sublattices with one occupied by Co$^{3+}$ ions and the other by Co$^{4+}$ ions. We have also measured the c-axis AMR for Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$ single crystals, and found no evidence for the existence of two- and four-fold symmetries.

The phase diagram of Na$_{x}$CoO$_2$ shows that this cobalt oxide is a paramagnetic metal at $x \sim 0.35$, a Curie-Weisse metal around $x \sim 0.66$, and a magnetically ordered metal at $x \sim 0.85$. However, at about $x = 0.5$, Na$_x$CoO$_2$ becomes a charge-ordered insulator in low temperatures. It was reported that Na$_{0.5}$CoO$_2$ exhibits three successive phase transitions at 88 K, 53 K and 20 K, respectively. A charge ordered state is formed in low temperatures. It causes a metal-insulator transition at 53 K. Furthermore, a static magnetic order was observed by the $\mu$SR measurements below this metal-insulator transition temperature. A weak magnetic transition was observed in the resistivity as well as low field (a few gauss) magnetic susceptibility measurements at 25 K. The $\mu$SR experiments also showed that there are two characteristic frequencies below 20 K, indicating the formation of two distinct types of local moments.

Recently, the c-axis magnetoresistance of Na$_{0.5}$CoO$_2$ was measured. It was found that a magnetic field applied along the c-axis direction has almost no effect on the metal-insulator transition temperature up to 43 Tesla. On the contrary, if a magnetic field is applied along the Co-O planes, the charge ordering is suppressed and the metal-insulator transition disappears above 40 Tesla. The c-axis AMR measurements indicates that the electronic structure changes from a two-fold to a six-fold symmetry when the magnetic field rotating within the Co-O planes is increased from 25 T to 45 T at 0.6 K.

In this paper, we report the measurement of the c-axis AMR for Na$_{0.48}$CoO$_2$ single crystal, with a rotating magnetic field of 10 Tesla applied within Co-O planes. As will be shown later, the AMR measurement is a useful tool for analyzing electronic states of materials with layered structures. It can reveal, for example, the symmetry of the Fermi surface as well as the distribution of electronically active ions.

The Na$_{0.48}$CoO$_2$ sample is prepared by sodium deintercalation from a Na$_{0.85}$CoO$_2$ single phased single crystal, synthesized by the floating zoom method. The single crystal of Na$_{0.85}$CoO$_2$ was marinated in a Br$_2$ and CH$_3$CN mixture solution for more than two weeks for sodium deintercalation. Sodium concentration determined by ICP is 0.48 ± 0.02. X-ray measurements show that Na$_{0.48}$CoO$_2$ is in a single phase and the c-axis lattice constant $c$ is about 11.04 A. The magnetoresistance is measured using a standard four-probe low frequency ac method in a PPMS system of Quantum Design company.

Figure 1(a) shows the c-axis resistance $R$ of the Na$_{0.48}$CoO$_2$ single crystal as a function of temperature $T$. The overall $R$–$T$ curve behaves similarly as for Na$_{0.5}$CoO$_2$. With decreasing temperature, $R$ increases gradually from 300 K to 40 K. It then shows a dramatic increase below 40 K, as a result of metal-insulator phase transition. Here the metal-insulator transition temperature is lower than the corresponding transition temperature for Na$_{0.5}$CoO$_2$. This difference is due to the differ-
ence in the Na concentration.

Figure 1(b) compares the temperature dependence of \( R \) at zero field with that in an external magnetic field of 10 T applied along the Co-O planes. It shows that the metal-insulator transition temperature is not changed by 10 T applied within the Co-O planes (solid squares), respectively.

\[ \text{FIG. 1: (a) Temperature dependence of the c-axis resistivity } R \text{ of the Na}_{0.48}\text{CoO}_2 \text{ single crystal. (b) } R \text{ versus } T \text{ below 50 K at 0 T (open triangles) and 10T applied within the Co-O planes (solid squares), respectively.} \]

Figure 2 shows the c-axis AMR for Na\(_{0.48}\)CoO\(_2\) at different temperatures. In low temperatures, the AMR shows a clear periodical oscillation decorated with fine structures with the change of the rotating angle \( \phi \). However, in high temperatures, some of these fine structures disappear.

\[ \text{FIG. 2: Angular dependence of the c-axis resistance of Na}_{0.48}\text{CoO}_2 \text{ single crystal at different temperatures in an applied magnetic field of 10T rotating within Co-O planes.} \]

In order to find the hidden periodicities associated with the fine structures of the AMR, we perform a Fourier analysis on the experimental data. The Fourier transformation of \( R(\phi) \) is defined by

\[ F_n = \frac{1}{N} \sum_{j=0}^{N-1} R(\phi_j) e^{-i\phi_j n} \]  

(1)

where \( \phi_j = 2\pi j/N \) and \( n = 0, 1, \ldots, N - 1 \). \( N \sim 450 \) is the total number of data points measured at each circle of rotation of the magnetic field. \( F_n \) is the \( n \)th coefficient of the Fourier expansion. Its absolute value \( |F_n| \) measures the contribution of the conduction channel with \( n \)-fold rotational symmetry.

\[ \text{FIG. 3: The Fourier power spectra of the c-axis AMR for Na}_{0.48}\text{CoO}_2 \text{ at different temperatures.} \]

Among them, the two-fold symmetric component \( (n = 2) \) is the most distinctive one. This indicates that the two-fold symmetric channel contributes most to the c-axis magnetoresistance below 50 K. This is apparently due to the formation of charge ordering in this temperature regime. In Na\(_x\)CoO\(_2\) there are both Co\(^{3+}\) and Co\(^{4+}\) ions. In the charge ordered state, it was believed that Co\(^{3+}\) and Co\(^{4+}\) ions will order independently to form alternating charge stripes.\(^{3,6}\) These charge stripes reduce the symmetry of the material and lead to the \( F_2 \) term in the c-axis AMR.
Similar two-fold symmetry has been observed in the in-plane as well as out-plane AMR in lightly doped high-$T_c$ cuprates $La_{2-x}Sr_xCuO_4$. In that material, the two-fold symmetry is believed to be due to the formation of charge stripes. The six-fold symmetry in AMR reflects naturally the triangular lattice symmetry of Na$_2$CoO$_2$. It is also a manifestation of the six-fold rotational symmetry of the Fermi surface of Na$_2$CoO$_2$ as revealed by the Angle-Resolved Photoemission measurements.\textsuperscript{16,17}

The observation of the distinctive four-fold symmetric component in low temperatures indicates that there is a square-lattice-like symmetry in Na$_{0.48}$CoO$_2$. In Na$_{0.5}$CoO$_2$, neutron and electron diffraction experiments revealed that Na atoms can form an orthorhombic sub-lattice with one occupied by Co$^{3+}$ ions and the other by Co$^{4+}$ ions with antiferromagnetic ordering. The Co$^{4+}$ ions, each of which contains a local moment with $S=1/2$, form an antiferromagnetic sub-lattice with dimension $a \times \sqrt{3}a$. The $\mu$SR and magnetic susceptibility experiments also suggest that there exists a spin long range order below 53 K, and this spin long range order may be antiferromagnetic correlated.\textsuperscript{6,10} The observation of the four-fold rotational symmetry in the AMR lends support to this picture.

Figure 5 shows the temperature dependence of $|F_2|$, $|F_4|$ and $|F_6|$. $|F_2|$ decreases with increasing temperature (\~{}40K). This suggests that the two-fold symmetry in the AMR is indeed due to the charge ordering in Na$_{0.48}$CoO$_2$. With increasing temperature, both $|F_4|$ and $|F_6|$ drop quickly in low temperatures and then merge into the noise background at about 25 K.

It is interesting to compare the AMR results for Na$_{0.48}$CoO$_2$ with the compounds that do not show any charge ordering phase in low temperatures. Figure 6 shows the c-axis AMR results for Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$ single crystals. In contrast to Na$_{0.48}$CoO$_2$, the c-axis AMR for Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$ does not show any periodical oscillations above the noise background. It is not surprising that the two- and four-fold symmetric components are absent in the AMR, since there is no charge ordering of Co and Na ions in both Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$. However, the absence of six-fold symmetry is probably due to the fact that the c-axis AMR in Na$_{0.35}$CoO$_2$ and Na$_{0.85}$CoO$_2$ are significantly smaller than that of Na$_{0.48}$CoO$_2$.

In summary, we have measured the c-axis AMR of a Na$_{0.48}$CoO$_2$ single crystal. We find that the oscillation of the AMR is dominated by a two-fold rotational symmetry below the metal-insulator transition temperature. This is consistent with the observation that the metal-insulator transition is induced by the charge ordering of Na atoms in the Co-O layers. As revealed by the Angle-Resolved Photoemission measurements, the charge ordering state of Na atoms can lead to an ordering state of charge carriers in the Co-O layers.\textsuperscript{16} Recently, Choy et al. proposed an insulating ground state of Na$_{0.5}$CoO$_2$ based on the calculation of the Hubbard model. As shown in figure 4, the triangular Co lattice is broken into two orthorhombic sublattices with one occupied by Co$^{3+}$ ions and the other by Co$^{4+}$ ions.\textsuperscript{12} The Co$^{4+}$ ions, each of which contains a local moment with $S=1/2$, form an antiferromagnetic sub-lattice with dimension $a \times \sqrt{3}a$. The $\mu$SR and magnetic susceptibility experiments also suggest that there exists a spin long range order below 53 K, and this spin long range order may be antiferromagnetic correlated.\textsuperscript{6,10} The observation of the four-fold rotational symmetry in the AMR lends support to this picture.

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It is interesting to compare the AMR results for Na$_{0.48}$CoO$_2$ with the compounds that do not show any
Co ions. The amplitudes of the oscillation in the AMR corresponding to the four- and six-fold rotational symmetries are also distinctive, but disappear at about 25 K. The six-fold oscillation reflects naturally the triangular lattice symmetry. The four-fold oscillation is speculated to result from the orthorhombic redistribution of Co ions in low temperatures.

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