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ScienceDirect
Journal of Magnetism and Magnetic Materials 310 (2007) 1027–1029
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

# $Ca_3Ru_2O_7$: Electronic instability and extremely strong quasiparticle renormalisation

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Available online 3 November 2006

**Abstract**

We report on the electronic structure of the bilayer ruthenate $Ca_3Ru_2O_7$ using high quality single-crystals grown by a floating-zone method which have the lowest residual resistivity so far achieved. The quantum oscillation, specific heat, and angle-resolved photoemission-spectroscopy (ARPES) measurements performed on these crystals establish that $Ca_3Ru_2O_7$ is a quasi two-dimensional low-carrier metal with a density-wave instability.


*PACS:* 74.70.Pq; 71.18.+y; 72.15.Gd

*Keywords:* $Ca_3Ru_2O_7$; Electronic structure; Transport properties; ARPES; Crystal growth

## 1. Introduction

The layered perovskite ruthenates have generated a wealth of novel physics since the discovery of superconductivity in $Sr_2RuO_4$ [1]. The superconducting order parameter has been shown to be unconventional [2], and extensive investigations of related compounds such as $Ca_2RuO_4$, $SrRuO_3$, $Sr_3Ru_2O_7$ and $Sr_4Ru_3O_{10}$ have revealed a number of novel phase transitions and magnetic states [3–7]. Among the series, the properties of the bilayer material $Ca_3Ru_2O_7$ have presented something of a mystery. Single crystals have been grown by both the flux method [8] and in image furnaces [9]. The measured resistivity ($\rho$) shows metallic ($d\rho/dT>0$) behaviour at low temperatures with a residual resistivity of 400 $\mu\Omega$ cm in Ref. [9], but flux-grown crystals present a non-metallic temperature dependence ($d\rho/dT<0$) [8]. Besides, the electronic specific heat ($\gamma$) has been reported to have a metallic component that varies considerably, from 1.7 $mJ/K^2$mol-Ru [9] to 20 $mJ/K^2$ mol-Ru [10]. Shubnikov–de Haas oscillations have been seen by both groups, but the only direct comparisons of the specific heat coefficient with the quasiparticle masses gave very poor agreement [11], leaving many open questions about the precise nature of the ground state of the material. In this paper, we describe the growth, in an image furnace, of metallic single crystals of $Ca_3Ru_2O_7$ with a residual resistivity one order of magnitude lowerthan the best previously observed value. Using these crystals, we have performed detailed electronic structure studies by means of quantum oscillations and high-resolution angle-resolved photoemission-spectroscopy (ARPES).

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doi:10.1016/j.jmmm.2006.10.225

## 2. Experiment

The $Ca_3Ru_2O_7$ crystals were grown by a floating-zone method with $RuO_2$-rich feed rods. The optimised feed rods for our image furnace were prepared with a composition of $Ca_2Ru_nO_y$ ($n = 2$) reflecting a heavy evaporation of $RuO_2$ during the crystal growth. The growth was done at the speed of 7 mm/h and under a pressure of 1 MPa with a flowing gas mixture ($Ar : O_2 = 9 : 1$). Crystals with the typical size of 3 mm × 4 mm × 1 mm (*c*axis) were obtained. The crystals were characterised by X-ray and SEM (scanning electron microscope), and the lattice parameter was deduced as $a = 0.5368$ nm, $b = 0.5522$ nm, and $c = 1.9565$ nm at room temperature, in qualitative agreement with previous reports [9,11,12]. The quantum oscillation (Shubnikov–de Haas effect), magnetoresistance, Hall resistivity, specific heat and angle-resolved photoemission spectroscopy measurements were performed using these as-grown crystals.

## 3. Transport properties and electronic structure

The in-plane resistivity $\rho_{ab}$ of two of the resulting crystals is plotted in Fig. 1 as a function of temperature down to 4 K. The most prominent features are associated with the previously-established Néel and first order structural phase transitions denoted as $T_N$ and $T_S$ [8,9,12,13]. At temperatures above these transitions, $\rho_{ab}$ is almost linear in $T$.

Approximately below 30 K, $\rho_{ab}$ drops dramatically, giving evidence for a strongly metallic temperature dependence. Using these metallic samples, measurements of the low temperature Hall effect, Shubnikhov–de Haas effect and magnetoresistance [14] show that in this regime the carrier concentration is extremely small ($\sim 1.4 \times 10^{-3}$ free carriers per Ru), assuming a quasi two-dimensional electronic structure, and hence that the best residual resistivity $\rho_{ab0} = 40\,\mu\Omega$ cm corresponds to a fairly long mean free path of 500 Å.

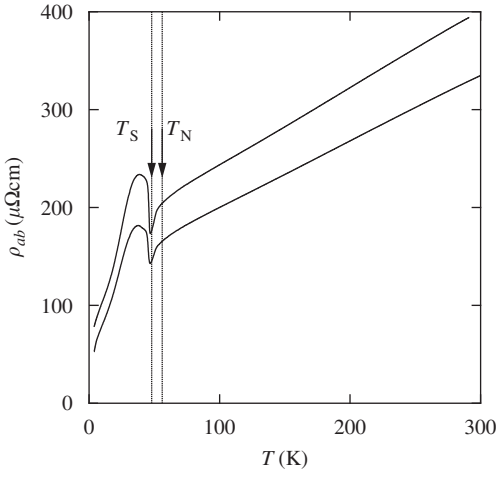


Fig. 1. In-plane resistivities of two representative $Ca_3Ru_2O_7$ crystals grown by floating-zone method, shown as a function of temperature. Indicated are the observed Néel ($T_N$) and structural transition temperatures ($T_S$).

By combining the two techniques of quantum oscillations and ARPES we are able to investigate the subtle electronic structure of this material in great detail [15]. We show that $Ca_3Ru_2O_7$ is a metal at low temperatures, with a Fermi surface consisting of two pockets which are quasi two-dimensional in character and have tiny volumes of approximately 0.3% of that of the first Brillouin zone. The ARPES study indicates that these pockets are due to a gap in the electronic band structure which opens gradually and coincides with the structural phase transition. Due to this electronic instability most of the Fermi surface is gapped away from the Fermi energy, leaving tiny pockets of carriers near the M and M′ points of the reduced Brillouin zone.

Intriguingly, the residual itinerant electron fluid is highly unusual. In fact, $Ca_3Ru_2O_7$ contains the lowest quasi particle residue $Z$ of Landau quasiparticles ever observed by ARPES [15]. Taking into account that the $Ca_3Ru_2O_7$ crystal structure has an orthorhombic symmetry, we developed a simple tight binding model for the band structure at the Fermi surface. Numerical calculations based on this model are able to capture the essential features of the topography of the electronic structure [14], but leave the origin of the renormalisation as an open question. A number of mysteries remain, for example the fact that the gap opens gradually while the structural transition is first-order.

This and other work [16] suggests that $Ca_3Ru_2O_7$ will be another "benchmark" material for the investigation of the complex interplay between lattice, orbital, spin and charge degrees of freedom that is a ubiquitous feature of transition metal oxide physics.

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