Second-order Structural Transition in 
(Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$

Y W Cheung$^1$, Y J Hu$^1$, S K Goh$^{1,*}$, K Kaneko$^2$, S Tsutsui$^3$, 
P W Logg$^4$, F M Grosche$^4$, H Kanagawa$^5$, Y Tanioku$^5$, M Imai$^5$, 
T Matsumoto$^5$, K Yoshimura$^5$

$^1$ Department of Physics, The Chinese University of Hong Kong, Shatin N.T., Hong Kong, China
$^2$ Materials Sciences Research Center, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki 319-1195, Japan
$^3$ Japan Synchrotron Radiation Research Institute (JASRI), SPring-8, Sayo, Hyogo 679-5198, Japan
$^4$ Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom
$^5$ Department of Chemistry, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan

E-mail: skgoh@phy.cuhk.edu.hk

Abstract. (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ is a member of the substitution series (Ca$_x$Sr$_{1-x}$)$_3$Rh$_4$Sn$_{13}$ which has recently been argued to feature a structural quantum critical point at $x_c$ = 0.9. In the stoichiometric compound Sr$_3$Rh$_4$Sn$_{13}$, the structural transition at $T^*$$\approx$ 138 K has been shown to be second-order. Moving towards $x_c$, we examine the character of the structural transition in (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ (i.e. $x$ = 0.5, $T^*$=$\approx$ 55 K) using electrical resistivity, heat capacity and X-ray scattering. The absence of the thermal hysteresis in specific heat around $T^*$, and the continuous evolution of the superlattice reflection detected by X-ray diffraction are consistent with the scenario that the structural transition associated with a modulation vector $q$ = (0.5 0.5 0) in (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ remains second-order on approaching the quantum critical point.

1. Introduction

Sr$_3$Rh$_4$Sn$_{13}$ is a conventional s-wave superconductor with a critical transition temperature ($T_c$) of 4.7 K [1, 2, 3]. In addition to the superconducting transition, another second-order phase transition takes place at $T^*$$=$138 K. This high temperature transition has been established to be a structural phase transition between the $Pm3n$ and $I43d$ space groups above and below $T^*$, respectively. $T^*$ can be suppressed either by applying pressure or by calcium substitution, i.e. by forming a (Ca$_x$Sr$_{1-x}$)$_3$Rh$_4$Sn$_{13}$ substitution series [1, 3]. If the structural transition remains second order as $T^*$$\rightarrow$ 0, a structural quantum critical point can be realized [1, 4]. In the substitution series (Ca$_x$Sr$_{1-x}$)$_3$Rh$_4$Sn$_{13}$, $T^*$ can be completely suppressed solely by fine tuning the calcium content, and it has been established that $T^*$$\rightarrow$ 0 when $x$ = $x_c$ $\approx$ 0.9 [1]. In the vicinity of $x_c$, several important observations are noted: (a) Debye temperature is a minimum, (b) $T_c$ is a maximum, and (c) strong-coupling superconductivity can be stabilized. All these features can be nicely understood in the framework of structural quantum criticality, and
they thus provide strong support for the identification of $x_c$ as a quantum critical point [1, 3]. In Sr$_3$Rh$_4$Sn$_{13}$ ($x=0$), specific heat data unambiguously proved that the structural transition is second-order [1, 3]. In this article, we move closer towards $x_c$, and demonstrate that the $T^*$ transition remains second-order in (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ ($x = 0.5$) using our latest x-ray diffraction data.

2. Experimental Details

Single crystals of (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ were grown using the Sn-flux method as described elsewhere [5]. Electrical resistivity and heat capacity were measured using Quantum Design’s Physical Properties Measurement System. High pressure electrical resistivity was measured using a piston-cylinder pressure cell with Daphne 7373 as the pressure transmitting medium. Superlattice reflection was measured at BL35XU of SPring-8, Japan with a Si(11 2 0 11) backscattering analyzer on the two-theta arm, which helps to reduce the background. The X-ray energy is 21.747 keV.

3. Results and Discussion

The inset to Figure 1 displays the universal temperature-pressure phase diagram constructed earlier [1], with the arrow marking the position of (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$. It is therefore more than halfway between $x = 0$ and $x_c$. The main panel of Figure 1 shows the temperature dependence of the electrical resistivity for (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ at ambient pressure, 6.0 kbar, 14.0 kbar and 20.6 kbar. In addition to the superconducting transition at low temperature, the structural transition at $T^* \sim 55$ K can be identified as the hump in the electrical resistivity. When pressure is applied, $T^*$ decreases rapidly, reaching 46.9 K and 29.7 K at 6.0 kbar and 14.0 kbar, respectively. At 20.6 kbar, the $T^*$ feature is no longer visible in the electrical resistivity. This confirms the trend that pressure moves the system towards the right hand side of the phase diagram.

In Sr$_3$Rh$_4$Sn$_{13}$, a pronounced lambda-like jump was detected at $T^*$ [1, 6], without any thermal hysteresis [1]. In the inset to Figure 2, we display the specific heat data of (Ca$_{0.5}$Sr$_{0.5}$)$_3$Rh$_4$Sn$_{13}$ around $T^*$ collected on warming and on cooling. The specific heat anomaly is significantly weaker here compared with the case of Sr$_3$Rh$_4$Sn$_{13}$. However, the absence of thermal hysteresis
is still clear. In the main panel of Figure 2, we present the temperature evolution of the elastic X-ray scattering intensity for \( Q = (6.5 \ 6.5 \ 1.0) \). The additional reflection at \( Q = k + q \) below \( T^* \), where \( k = (6.0 \ 6.0 \ 1.0) \) corresponds to the Bragg spots in the high temperature (\( T > T^* \)) phase, is consistent with a modulation vector with \( q = (0.5 \ 0.5 \ 0.0) \) (or the M point). \( T^* \) determined from the onset of the superlattice reflection is slightly below that obtained from the specific heat data. This difference can be attributed to a slight variation in the calcium content between different samples. From the slope \( dT^*/dx \) (c.f. Figure 1 and Ref. [1]), a variation of 0.03 in \( x \) can cause a variation of up to 5 K in \( T^* \). Finally, the gradual and continuous increase of the diffraction intensities on cooling below \( T^* \) is consistent with the scenario of a second-order phase transition.

4. Conclusion

In summary, we have studied single crystals of \((\text{Ca}_{0.5}\text{Sr}_{0.5})_3\text{Rh}_4\text{Sn}_{13}\) using electrical resistivity, heat capacity and X-ray diffraction. From the absence of the thermal hysteresis around \( T^* \) in the heat capacity data and the continuous evolution of the superlattice peak intensity at (6.5 6.5 1), we conclude that the structural transition in \((\text{Ca}_{0.5}\text{Sr}_{0.5})_3\text{Rh}_4\text{Sn}_{13}\) is a second-order phase transition with \( q = (0.5 \ 0.5 \ 0.0) \). Similar conclusion has recently been reached in related compounds \(\text{La}_3\text{Co}_4\text{Sn}_{13}\) [7], \(\text{Sr}_3\text{Ir}_4\text{Sn}_{13}\) [6], \(\text{Sr}_3\text{Rh}_4\text{Sn}_{13}\) [6] and \(\text{Ca}_3\text{Ir}_4\text{Sn}_{13}\) [8] from x-ray/neutron diffraction. These data highlights the similarity across different 3-4-13 families in which the notion of structural quantum criticality has been discussed [1, 4, 9].

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