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Pressure effects on the structural and superconducting transitions in $La_3Co_4Sn_{13}$

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ABSTRACT

$La_3Co_4Sn_{13}$ is a superconducting material with transition temperature at $T_c = 2.70$ K, which presents a superlattice structural transition at $T^* \simeq 150$ K, a common feature for this class of compounds. However, for this material, it is not clear that at T^* the lattice distortions arise from a charge density wave (CDW) or from a distinct microscopic origin. Interestingly, it has been suggested in isostructural non-magnetic intermetallic compounds that T^* can be suppressed to zero temperature, by combining chemical and external pressure, and a quantum critical point is argued to be observed near these critical doping/pressure. Our study shows that application of pressure on single-crystalline $La_3Co_4Sn_{13}$ enhances T_c and decreases T^*. We observe thermal hysteresis loops for cooling/heating cycles around T^* for $P \gtrsim 0.6$ GPa, in electrical resistivity measurements, which are not seen in x-ray diffraction data. The hysteresis in electrical measurements may be due to the pinning of the CDW phase to impurities/defects, while the superlattice structural transition maintains its ambient pressure second-order transition nature under pressure. From our experiments we estimate that T^* vanishes at around 5.5 GPa, though no quantum critical behavior is observed up to 2.53 GPa.

1. Introduction

There has been a revived interest in the $R_3M_4X_{13}$ (3-4-13) (R = rare-earth or alkaline-earth element; M = transition metal and X = groups-13,14 element) compounds due to their diverse physical properties, which include antiferromagnetic [1–4], superconducting [4,5], strong electronic correlations [4,6] and semiconducting [5,7] behavior. The crystal structure of this 3-4-13 series, at room temperature, is the cubic $Yb_3Rh_4Sn_{13}$ type structure ($Pm\bar{3}n$ space group) [8]. The cubo-octahedral R site has a small distortion, resulting in a local tetragonal symmetry [9]. For M = Co and X = Sn, all known compounds undergo a superlattice structural transition at a temperature T^*, which doubles the lattice parameter in respect to the higher temperature phase [9], with propagation vector of $\mathbf{q} = \{(0.5\ 0.5\ 0), (0.5\ 0\ 0.5), (0\ 0.5\ 0.5)\}$ [10]. This superlattice structural transition was also observed for the $Sr_3Ir_4Sn_{13}$ superconductor and the existence of a charge density wave (CDW) state below $T^* \simeq 147$ K was reported [11]. By combining Ca^{2+} substitution and application of external pressure P, T^* is suppressed to zero, suggesting a superlattice quantum phase transition at zero temperature [11], analogous to those near a magnetic [12,13], superconducting [14] and ferroelectric [15] instabilities. Later, a structural quantum critical point was also described in the $Sr_{3-x}Ca_xRh_4Sn_{13}$ system at ambient pressure [16].

Due to its similarity with the $Sr_{3-x}Ca_x(Rh,Ir)_4Sn_{13}$ series of materials, the $La_3Co_4Sn_{13}$ compound, presenting a superconducting transition temperature at $T_c = 2.70$ K, has recently attracted attention [17–19]. The superconducting state is argued to be *s*-wave, in the strong-coupling regime [20,21], such as others 3-4-13

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superconductors [22–24]. The superlattice structural transition of $La_3Co_4Sn_{13}$ occurs at $T^* \simeq 150$ K and whether this transition is of first or second-order is still under debate [18,19]. It is also controversial whether at T^* the lattice distortions arise from a conventional CDW [18,25].

Pressure effects studies on superconductors are of great interest since it gives insights on the microscopic superconducting mechanism and may also induce interesting physical phenomena that might arise out of ambient conditions. For simple metal superconductors it is generally expected that T_c decreases with pressure. This is because the density of states at the Fermi energy [$N(E_f)$] and the effective attractive pairing interaction (V_{eff}), which are both related to T_c through $T_c \propto e^{-1/[N(E_f)V_{eff}]}$, vary with pressure. While $N(E_f)$ usually decreases with the reduction of the unit cell volume, V_{eff} is significantly affected due to the difficulty for the crystal lattice to couple with the electrons [26,27]. Oddly, it was observed that for a polycrystalline $La_3Co_4Sn_{13}$ sample ($T_c = 1.95$ K at ambient pressure) T_c increases linearly with applied pressure at a rate of $dT_c/dP \sim 0.03$ K/GPa [28], in contrast to M = Rh [28] and Ru [29] in which T_c decreases with pressure. An increase of T_c up to 5.1 K, for 10% In substitution, was also reported, though application of pressure in $La_3Co_4Sn_{11.7}In_{1.3}$ decreased T_c by −0.3 K/GPa [30]. While the enhancement of T_c against pressure is attributed to the decrease of the structural instability [28], the results for In substitution, based on theoretical calculations, is argued to be related to modifications in band structure and density of states [30].

In this work we present our investigation of pressure effects on single-crystalline superconducting $La_3Co_4Sn_{13}$ compound, via electrical resistivity and x-ray diffraction. We observe a positive variance of T_c under pressure, with a faster rate than for polycrystalline samples. Additionally, a thermal hysteresis loop around the superlattice structural transition is seen in electrical resistivity experiments for $P \gtrsim 0.6$ GPa, presumably due to pinning of a partially gapped CDW phase, which sets in at T^*. This feature was not previously reported, giving evidence for a concomitant CDW at T^*, and may be a common characteristic of some 3-4-13 materials. We also show the decrease of this second-order superlattice structural transition with pressure and estimate that it vanishes at $P \sim 5.5$ GPa, however, with no indication of quantum critical behavior up to $P = 2.53$ GPa.

2. Experimental details

Flux-grown single crystals of $La_3Co_4Sn_{13}$ were synthesized using Sn excess [17]. Phase purity was checked by x-ray diffraction (XRD) on powdered crystals (not shown). Ambient pressure synchrotron XRD data ($E = 8.33$ keV) at cooling and heating cycles were collected for a single crystalline sample ($\sim 2 \times 1 \times 1$ mm^3), cut and polished to achieve a flat and shiny surface perpendicular to the [110] direction at beamline P09 at PETRA III (DESY/Germany) [31]. Temperature dependent DC electrical resistivity measurements were performed in a Quantum Design DynaCool PPMS, by means of the conventional four-contact configuration. For experiments under hydrostatic pressure, several samples were screened for Sn inclusions. We used a clamp-type Cu-Be cell, with silicon oil as pressure transmitting medium. Lead was used as pressure manometer. Synchrotron XRD measurements under pressure were performed at beamline XDS at UVX (LNLS/Brazil) [32]. The sample/diamond anvil cell (DAC) was placed in the cold-finger of a He cryostat. The powder patterns were collected with an area detector (MAR225) and the two-dimensional (2D) images were integrated to provide intensity as a function of 2θ using the FIT2D software [33]. Due to the DAC limited angular scattering range (25° of scattering angle 2θ), the beam was tuned to $E = 20$ keV in order to detect a significant number of Bragg peaks. Hydrostatic pressure was generated using a DAC with diamond anvils of 900 μm culet size and silicon oil was used as pressure media. To calibrate *in situ* the pressure, ruby spheres were loaded with the fine powdered samples in the stainless steel gasket specimen chamber.

3. Results

Fig. 1 shows the temperature dependence, upon heating up and cooling down the sample, of the integrated intensity for the superstructure reflection (3.5 2 0.5) (at ambient pressure) of $La_3Co_4Sn_{13}$. A continuous decrease of the superstructure peak and no appreciable thermal hysteresis, within the instrument resolution, indicate a second-order phase transition at $T^* \simeq 150$ K, as in earlier reports [19,25]. In addition, the inset exhibits the rocking curves around the superstructure peak used to calculate the integrated intensities. In order to gain more insight into the physical properties of this structural distortion, the temperature dependent data was fitted by a power-law expression $[(T^* - T)/T^*]^{2\beta}$ yielding $T^* = 150.0(1)$ K and a critical exponent $\beta = 0.36(1)$. The critical exponent for our single crystal suggests a three-dimensional character of the structural distortion, in good agreement to what is observed on single crystalline $Sr_3Ir_4Sn_{13}$ [34].

The temperature dependent electrical resistivity $\rho(T)$ of our $La_3Co_4Sn_{13}$ single crystals, at ambient pressure, show metallic behavior down to low temperatures, until the superconducting transition takes place at $T_c \simeq 2.7$ K. Additionally, a small kink due to the superlattice structural transition is observed at $T^* \simeq 146$ K, consistent with our synchrotron XRD data. No thermal hysteresis was also observed for $\rho(T)$ and specific heat measurements at $P = 0$. These data are consistent with previous works [17,19] and are not shown here.

Representative curves of the pressure evolution of T_c in $La_3Co_4Sn_{13}$ for our $\rho(T)$ data are presented in Fig. 2 (a). A sharp transition near T_c is observed, which does not broaden with pressure. Similarly to polycrystalline $La_3Co_4Sn_{13}$, our single crystalline samples also exhibit an enhancement of T_c with pressure, increasing linearly at a rate of $dT_c/dP \sim 0.07(1)$ K/GPa [Fig. 3(b)], more than double of the polycrystalline material rate [28], but one order of magnitude lower than observed for the initial dT_c/dP slope

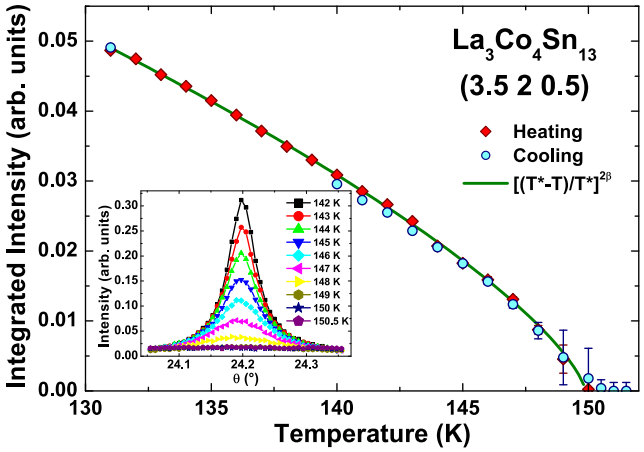

Fig. 1. Temperature dependence of the (3.5 2 0.5) superlattice reflection at ambient pressure for cooling (circles) and heating (diamonds) cycle. The green solid line is a fitting using a power law to determine T^*. The inset shows the rocking curves around the superlattice reflection measured for selected temperatures during the heating cycle. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)