Possible indicators for low dimensional superconductivity in the quasi-1D carbide Sc3CoC4

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Possible indicators for low dimensional superconductivity in the quasi-1D carbide Sc_3CoC_4

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Abstract. The transition metal carbide Sc_3CoC_4 consists of a quasi-one-dimensional (1D) structure with $[CoC_4]_\infty$ polyanionic chains embedded in a scandium matrix. At ambient temperatures Sc_3CoC_4 displays metallic behavior. At lower temperatures, however, charge density wave formation has been observed around 143 K which is followed by a structural phase transition at 72 K. Below $T_c^{onset} = 4.5$ K the polycrystalline sample becomes superconductive. From $H_{c1}(0)$ and $H_{c2}(0)$ values we could estimate the London penetration depth ($\lambda_L \cong 9750$ Å) and the Ginsburg-Landau (GL) coherence length ($\xi_{GL} \cong 187$ Å). The resulting GL-parameter ($\kappa \cong 52$) classifies Sc_3CoC_4 as a type II superconductor. Here we compare the puzzling superconducting features of Sc_3CoC_4, such as the unusual temperature dependence i) of the specific heat anomaly and ii) of the upper critical field $H_{c2}(T)$ at T_c, and iii) the magnetic hysteresis curve, with various related low dimensional superconductors: e.g., the quasi-1D superconductor $(SN)_x$ or the 2D transition-metal dichalcogenides. Our results identify Sc_3CoC_4 as a new candidate for a quasi-1D superconductor.

1. Introduction

Low dimensional superconductivity is a vital and controversial research topic in solid sate physics and chemistry. However, beside the well-established examples for quasi-one-dimensional (1D) superconductors, e. g., polysulfur nitride $(SN)_x$ [1], the organic Bechgaard salts [2] or the transition-metal trichalcogenides [3], there are only a few systems known which allow us to study the nature of the phenomenon in greater detail. Recently, we observed superconductivity in Sc_3CoC_4 which might represent a new benchmark system of a quasi-1D superconductor [4]. Sc_3CoC_4 consists of quasi-one-dimensional $[CoC_4]_\infty$ polyanionic chains embedded in a scandium matrix [5]. From specific heat, magnetic susceptibility, resistivity and X-ray diffraction measurements a charge density wave around 143 K and a structural phase transition below 72 K were identified [4]. Therefore, it is very natural to ask if the superconductivity of this quasi-1D carbide is also of low dimensional character. In the absence of a single crystal, we compare in this paper i) structural features and physical properties like ii) the specific heat anomaly of the superconducting transition, iii) the magnetization behavior, and iv) the temperature dependence of the upper critical field with the established low dimensional superconductors like polysulfur nitride $(SN)_x$, the Bechgaard salts, high-T_c-cuprates, and the dichalcogenide $NbSe_2$ [7].

2. Structural details

High resolution single crystal X-ray diffraction studies at room temperature provide precise structural parameters of Sc_3CoC_4 (space group Immm) [5]: the Co atoms are coordinated by four

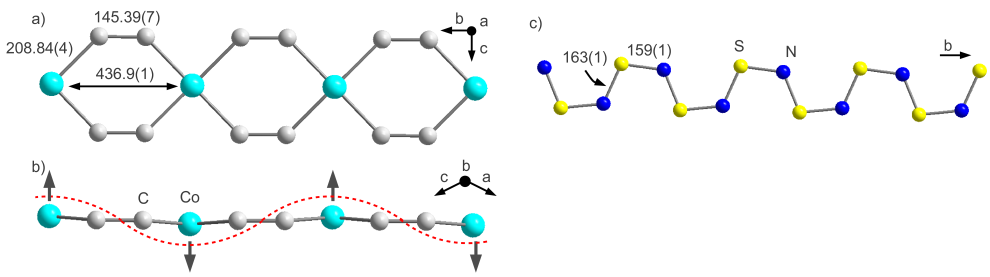

Figure 1. The $[CoC_4]_\infty$ ribbons of Sc_3CoC_4 a) in the crystallographic b,c plane and b) in the low temperature modification (side view) [4]. c) The $[SN]_\infty$ ribbons of the polysulfur nitride $(SN)_x$ showing a zigzag pattern with alternating short and long S-N distances [8]. Selected bond distances are given in pm.

C_2 pairs in an almost square planar manner, establishing one-dimensional infinite $[CoC_4]_\infty$ chains as pictured in Fig. 1a. Below the structural phase transition at 72 K Sc_3CoC_4 is characterized by an alternating displacement of the Co atoms above and below the $[CoC_4]_\infty$ ribbons (Fig. 1b, space group $C\,2/m$). Such a zigzag chain deformation pattern is also observed in quasi-1D superconductors like polysulfur nitride $(SN)_x$ (Fig. 1c)[8], or the organic Bechgaard salts [9]. Therefore, the zigzag distortion of the $[CoC_4]_\infty$ ribbons might be a structural prerequisite of the low-dimensional superconductivity, even though the zigzag pattern in Sc_3CoC_4 is only weakly pronounced in comparison to $(SN)_x$. However, for the non superconducting analogues Sc_3NiC_4 and Sc_3FeC_4 no structural phase transition was observed above 2 K [4].

3. Experimental results and discussion

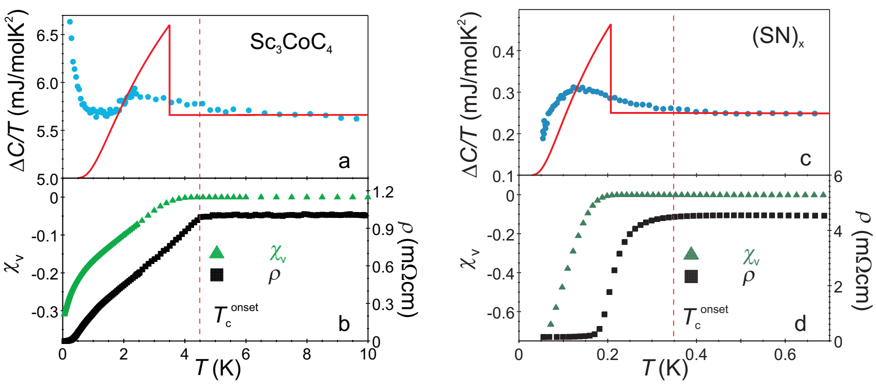

Figure 2. Temperature dependence of a) the electronic contribution of the specific heat divided by temperature, $\Delta C/T$, and b) the volume susceptibility, χ_V, at $H = 1$ Oe (left scale; triangles) and the electrical resistivity, ρ, (right scale; squares) for Sc_3CoC_4 in comparison with the temperature dependence of the same physical properties, c) $\Delta C/T$ and d) χ_V, ρ for the quasi-1D superconductor $(SN)_x$. The solid lines represent the superconducting transition as it is predicted by the BCS model [10].

The low temperature electronic specific heat of Sc_3CoC_4, shown as $\Delta C/T$ vs. T plot in Fig. 2a, reveals a broad superconducting transition below $T_c^{onset} = 4.5$ K which was determined by resistivity measurements (Fig. 2b). Additionally, this transition is indicated by the magnetic susceptibility showing a lowering of the magnetic response just below 4.4 K. These results and the fact, that the volume susceptibility at 50 mK approaches -0.3, identifies Sc_3CoC_4 as a bulk superconductor (Fig. 2b). The steep increase of $\Delta C/T$ below 1 K is mainly due to hyperfine interactions caused by the nuclear quadrupole moments of the Sc and Co atoms.

For the superconducting transition in Sc_3CoC_4 we calculated the expected ideal BCS curve of a weak coupled superconductor, taking into account the entropy balance between the normal and superconducting state (solid line in Fig. 2a) [10]. This calculation indicates, that only a small part ($\Delta\gamma_n = 0.65 \pm 0.05\,\mathrm{mJ/mol\,K^2}$) of the total electronic specific heat $\gamma_n = 5.7\,\mathrm{mJ/molK^2}$, which was graphically determined from a C/T vs. T^2 plot, can be involved in the development of the superconducting state. Therefore, the bulk superconductivity may be related solely to small areas of the Fermi-surface. Furthermore, the overall shape of the specific heat anomaly differs substantially from the BCS behavior, missing the characteristic pronounced jump at T_c.

The unusual broad shape of this anomaly and the fact, that only a small fraction (12 % in Sc_3CoC_4) of the band electrons contribute to the superconducting state is also observed in the well known quasi-1D superconductor polysulfur nitride $(SN)_x$ (Fig. 2c). Our specific heat measurement on a single crystal $(SN)_x$ sample indicates that 60 % of the total electronic specific heat ($\gamma_n = 0.25\,\mathrm{mJ/molK^2}$) is involved in the superconducting pairing process. The volume susceptibility at 70 mK approaches only a value of -0.66 of the full diamagnetic response (Fig. 2d). These analogies suggest that the unusual broad shape of the specific heat anomaly and the small percentage of band electrons contributing to the superconducting state may be characteristic features for the presence of quasi-one-dimensional superconductivity in Sc_3CoC_4.

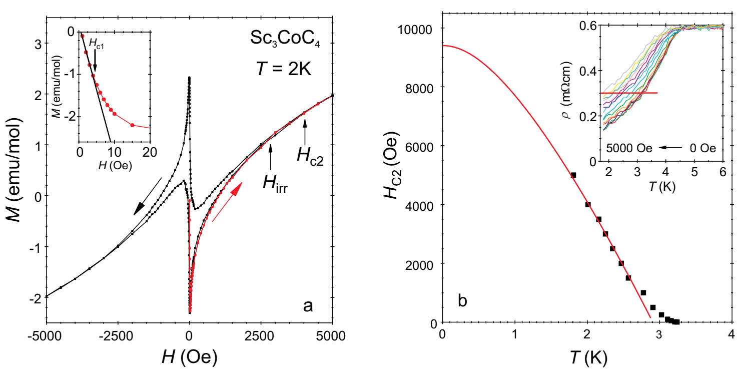

Figure 3. a) Magnetic hysteresis loop of Sc_3CoC_4 at 2 K. The underlying pronounced slope (M/H) and the deviation from the linear behavior of the magnetization is possibly due to the Pauli susceptibility of the normal conducting electrons and of a small impurity concentration of elementary Co, respectively. To determine H_{c1}, the insert displays an expanded region of the initial magnetization curve up to 20 Oe. b) Phase diagram of the upper critical fields $H_{c2}(T)$. The solid line corresponds to a WHH-fit [12]. $T_c(H_{c2})$ is obtained from the 50 % values of the normal state resistivity (solid line in the insert).

A further hint for low-dimensional superconductivity in Sc_3CoC_4 presents the magnetic hysteresis loop (Fig. 3a). This magnetization curve clearly indicates a type-II superconducting behavior. The characteristic dip near zero field crossings and the high reversibility between the irreversibility field ($H_{irr}(2K) = 2800 \pm 400\,\mathrm{Oe}$) and the upper critical field ($H_{c2}(2K) = 4000 \pm 200$) is also found in layered cuprate superconductors, but only at higher temperatures [6]. In Sc_3CoC_4 and also in the 2D highly anisotropic dichalcogenide superconductors like $NbSe_2$ [7, 11] such a reversible behavior above a threshold field occurs at very low temperatures and therefore might not only be due to granularity [6] but also due to the existence of low-dimensionality.

From $H_{c1} = 4.4\pm0.2$ Oe at 2 K, as determined by the deviation from the initial linearity of the magnetization curve (insert in Fig. 3a), the lower critical field $H_{c1}(0)$ is estimated to be 7 ± 0.4 Oe. $H_{c2}(0) = 94000\pm500$ is calculated from the WHH-theory [12], with a Maki parameter of $\alpha = 0.25$ estimated from the slope of the upper critical field at T_c and the spin-orbit parameter λ_{so} assumed to be zero. By combining the upper and lower critical field values with the Ginsburg Landau (GL) expressions for isotropic superconductors ($H_{c2} = \Phi_0/(2\pi\xi_{GL}^2)$; $H_{c1} = (\Phi_0/(4\pi\lambda_L^2))(0.08 + ln\kappa)$) we obtained the London penetration depth of $\lambda_L = 9750 \pm 300$ Å and the GL-coherence length of $\xi_{GL} = 187 \pm 5$ Å. The resulting GL parameter $\kappa = 52$ classifies Sc_3CoC_4 as a type-II superconductor.

A further interesting effect is the upturn of $H_{c2}(T)$ at temperatures close to T_c (Fig. 3b). Beside other explanations, e.g., sample imperfections, this effect is mainly attributed to a reduction of the dimensionality. This upturn is also found in the quasi-1D superconductors $(SN)_x$ [13], the Bechgaard salts [14] and as R. A. Klemm summarised: "seems to be a general feature of many, but not all highly anisotropic materials" [15].

4. Summary

Resistivity, magnetization and specific heat measurements clearly support the presence of bulk superconductivity in the quasi-one-dimensional transition metal carbide Sc_3CoC_4. Several characteristic features might identify Sc_3CoC_4 as a promising system in the rare class of quasi-one-dimensional superconductors: i) the structural zigzag chain, ii) the broad shape of the specific heat anomaly, iii) the small percentage of band electrons contributing to the superconductivity state, iv) the partially reversibility of the magnetic hysteresis loop, and v) the upturn of the $H_{c2}(T)$ at temperatures near T_c. However, future work on single crystalline samples of Sc_3CoC_4 is essential to analyze in detail the electronic anisotropy in both, the normal and the superconducting state.

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