Physica C: Superconductivity and its applications 528 (2016) 73–83

Contents lists available at ScienceDirect

Physica C: Superconductivity and its applications

journal homepage: www.elsevier.com/locate/physc

Physical properties and electronic structure of La_3Co and La_3Ni intermetallic superconductors

J. Strychalska[a,*], M. Roman[a], Z. Sobczak[a], B. Wiendlocha[b], M.J. Winiarski[a], F. Ronning[c], T. Klimczuk[a,*]

[a] Faculty of Applied Physics and Mathematics, Gdansk University of Technology, Narutowicza 11/12, 80-233 Gdansk, Poland
[b] Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, Aleja Mickiewicza 30, 30-059 Krakow, Poland
[c] Los Alamos National Laboratory, Los Alamos, New Mexico, 87545, USA

ARTICLE INFO

Article history:
Received 26 May 2016
Revised 15 July 2016
Accepted 23 July 2016
Available online 26 July 2016

Keywords:
Superconductivity
Intermetallic compounds
Band structure calculations

ABSTRACT

La_3Co and La_3Ni are reported superconductors with transition temperatures of 4.5 and 6 K, respectively. Here, we reinvestigate the physical properties of these two intermetallic compounds with magnetic susceptibility χ, specific heat C_p and electrical resistivity ρ measurements down to 1.9 K. Although bulk superconductivity is confirmed in La_3Co, as observed previously, only a trace of it is found in La_3Ni, indicating that the superconductivity in La_3Ni originates from an impurity phase. Superconducting state parameters for La_3Co, including lower and upper critical fields and the superconducting gap, are estimated. Results of the theoretical calculations of the electronic structure for both materials are also presented, and comparison of the Fermi level location in La_3Co versus La_3Ni explains its larger superconducting T_c. A major discrepancy between band structure calculations and the experimentally measured Sommerfeld coefficient is found. The measured electronic density of states is about 2.5 times larger than the theoretical value for La_3Co. This effect cannot be explained by the electron-phonon interaction alone. Renormalization of γ, as well as an $\sim T^2$ behavior of the resistivity, suggests the presence of spin fluctuations in both systems.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

RE_3M binary intermetallics, where *RE* is a rare earth metal and M = Co or Ni, belong to a large family of compounds which crystallize in the orthorhombic crystal structure with space group *Pnma* (s.g. #62, Pearson symbol *oP*16). A prototype of this structure is an iron carbide known as cementite with the chemical formula Fe_3C. Its characteristic feature are trigonal prisms that are formed by iron atoms and centered by a carbon atom (see Fig. 1). Cementite has been studied for decades and remains an important field of study [1,2].

Electrical and magnetic properties [3,4] as well as specific heat [5] of several RE_3Co binary compounds have been studied in details. Neutron diffraction studies on Dy_3Co [6] and Er_3M (M = Co, Ni) [7] show that only *RE* atoms carry a magnetic moment whereas Co and Ni atoms remain non-magnetic. Therefore replacing a magnetic lanthanide by lanthanum in RE_3M should lead to paramagnetic behavior and since there is a high concentration of La one can naively expect the presence of superconductivity. Indeed, C. S. Garde, et al. reported superconductivity in the La_3M series for various M metals including Ni and Co [8]. The highest superconducting critical temperature, estimated from resistivity measurements, was reported for La_3In ($T_c = 9.7$ K) and T_c of approximately 6 K was estimated for as many as four compounds: La_3Ga, La_3Sn, La_3Al and La_3Ni.

The most intriguing superconductors in the La_3M family are La_3Co and La_3Ni. Superconductivity in Co and Ni containing compounds is rather rare due to the strong magnetic character of Co and Ni atoms, that might break the Cooper pairs and destroy superconductivity. Fisk and Lawson reported superconductivity in La_3Co with $T_c = 4.3$ K [9], and more detailed studies, including electrical resistivity, magnetic susceptibility, heat capacity and thermopower, were performed later [10–12]. An intriguing feature of the resistivity data for La_3Co and La_3Ni is a $\rho(T) \sim AT^2$ dependence below 20 K, characteristic of Fermi-liquid behavior. From the resistivity and the heat capacity data, Lu, et al. calculated a Kadowaki-Woods ratio for La_3Co and obtained a value of $A/\gamma^2 = 0.46 \cdot 10^{-6}$ Ω m (K mol/J)^2 that is close to the "universal" value of 10^{-7} Ω m (K mol/J)^2 observed in heavy-fermion compounds [11]. A slightly lower value $A/\gamma^2 = 0.28 \cdot 10^{-6}$ Ω m (K mol/J)^2 was found for La_3Ni

* Corresponding authors.
E-mail addresses: jstrychalska@mif.pg.gda.pl (J. Strychalska), tomasz.klimczuk@pg.gda.pl (T. Klimczuk).

http://dx.doi.org/10.1016/j.physc.2016.07.017
0921-4534/© 2016 Elsevier B.V. All rights reserved.

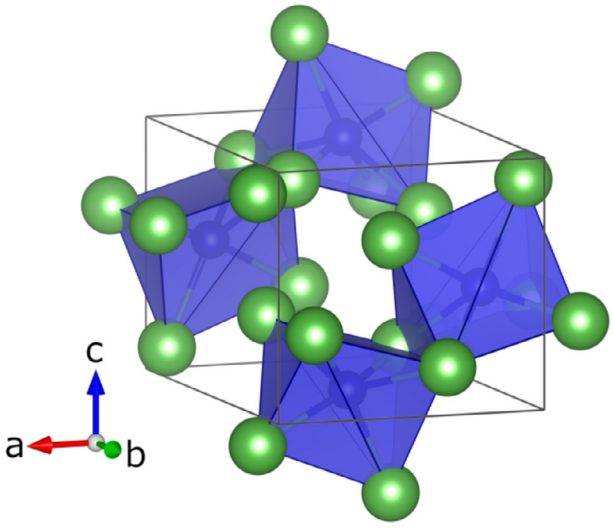

Fig. 1. Unit cell of cementite-type La_3Co intermetallic compound. Cobalt atoms (blue) at 4*c* site are surrounded by lanthanum atoms (green) from 8*d* and 4*c* positions, forming trigonal prisms.

[13]. The latter compound was claimed to be an unconventional superconductor with $T_c \sim 1.45$ K, based on heat capacity measurements [13,14]. However, a second heat capacity peak in samples that are nominally La_3Ni at $T \sim 2.4$ K [14] is likely caused by a La_7Ni_3 impurity phase [15], and, therefore, the unconventional character of La_3Ni is questionable.

The aim of this article is to present details of the superconducting properties of binary intermetallic La_3Co superconductor, compare them with La_3Ni, and analyze in view of the bandstructure calculations. Measurements of magnetization, specific heat and transport properties confirm bulk superconductivity of La_3Co, and in contrast only filamentary superconductivity is observed for La_3Ni.

The paper is structured as follows. In Section 2 the experimental details are provided. The electrical resistivity, magnetic susceptibility and heat capacity data for La_3Co and La_3Ni are given in Section 3.1. and Section 3.2, respectively. We provide superconducting parameters for La_3Co and compare the normal state parameters for La_3Co and La_3Ni. In Section 4 we discuss the band structure calculations for both compounds and we conclude in Section 5.

2. Experimental

Polycrystalline samples of La_3Co and La_3Ni were synthesized by arc-melting stoichiometric amounts of lanthanum (99.9%) and cobalt (99.95%) or nickel (99.95%), respectively. Melting took place under ultra-high purity argon atmosphere, in a water-cooled copper hearth. We used tungsten as the electrode material, and a zirconium button was used as an oxygen getter. The samples were remelted four times to get homogeneous materials. The arc-melted buttons were wrapped in tantalum foil, placed in evacuated quartz tubes and annealed at 500°C for two weeks. The phase purity of samples was checked by powder x-ray diffraction (XRD) on a PANalytical X'Pert Pro diffractometer with a Cu K_α source. PowderCell software was used for XRD pattern simulation [16].

All measurements of physical properties were performed by using a Quantum Design Physical Properties Measuring System PPMS-9. For La_3Co the isothermal magnetization and temperature dependence of the *dc* magnetic susceptibility were measured. Furthermore, the temperature dependence of the *ac* magnetization was measured on both compounds in a *dc* field of 5 Oe and 1 kHz excitations with an amplitude of 3 Oe. The specific heat measurements were performed using a standard relaxation method between 1.9 and 300 K at 0 and 3 T magnetic field. The electrical resistivity measurements were carried out by the standard four-probe method with 50 μm diameter platinum wires spot-welded to the samples.

3. Results

3.1. Powder x-ray diffraction

XRD patterns measured on powdered samples are shown in Supplementary material Fig. S1 and Fig. S2. As both samples oxidize quickly in air only short XRD scans could be performed. The obtained XRD patterns match the diffraction patterns of La_3Co and La_3Ni (Refs. [8] and [17,18] respectively). A large number of observed diffraction reflections results from low symmetry of the cementite-type structure. There are some reflections from unrecognized phase observed for both samples, most probably arising from minor amounts of β-lanthanum or lanthanum oxides.

3.2. Physical properties of La_3Co

The temperature dependence of the normal-state resistivity $\rho(T)$ of La_3Co between 1.9 K and 300 K at $H=0$ is presented in the main panel of Fig. 2. The resistivity data display metallic-like character ($dp/dT > 0$). The residual resistivity (ρ_0) estimated at $T=4.7$ K before entering the superconducting state is 43 μΩ cm and the residual resistivity ratio ($RRR \equiv \rho_{300\ K}/\rho_0$) is 6. An inset of Fig. 2 presents the zero-field-cooled (ZFC) and field-cooled (FC) *dc* volume magnetic susceptibility data $\chi(T)$ in the vicinity of the superconducting transition. The diamagnetic response was normalized by a demagnetization factor that was estimated from isothermal magnetization measurements M(H) in the superconducting state and will be described later. The superconducting critical temperature (T_c) was taken as the temperature where the extrapolation of the normal state susceptibility to lower temperatures intersects the extrapolation of the steepest slope of the $\chi(T)$ curve [19]. The T_c found for La_3Co in these measurements is 4.3 K, which is close to the previously reported values [9]. The much weaker FC signal indicates a significant pinning effect on grain boundaries.

To highlight the superconducting transition, the low temperature resistivity curves $\rho(T)$, under applied magnetic fields from 0 T to 3 T with a step of 0.2 T, are shown in the main panel of Fig. 3. In the zero-field data, a sudden drop of resistivity is observed below ~6 K, but the transition is not complete. Another drop is seen below 5 K, finally reaching a zero resistance state at 4.6 K. This behavior is likely caused by a small fraction of unreacted β-lanthanum (fcc type) with $T_c = 5.95$ K [20]. β–La has a critical field of ~0.11 T [21] and therefore at higher fields the measured superconducting transition becomes sharp and is representative of La_3Co with a width of 10 mK estimated for $\mu_0 H = 0.6$ T. Above 2 T, a two step transition is seen again, and for the highest magnetic field applied (3T) $T_{c\ onset}$ is about 2.3 K (marked by a vertical arrow). The origin of this behavior is unclear, although, it might originate from the presence of an unknown impurity phase, which superconducts with a rather large upper critical field. From our $\rho(T)$ measurement we conclude that the zero-field T_c for La_3Co is just above 4.6 K, the temperature at which the transition is complete.

In the inset of Fig. 3, the upper critical field determined from the resistivity measurements for La_3Co is plotted. The orbital critical field, for a one-band BCS-type superconductor, can be calculated from the Werthamer-Helfand-Hohenberg expression: $H_{c2}(T) = -AT_c \frac{dH_{c2}}{dT}|_{(T=T_c)}$ where A is 0.69 and 0.73 in the dirty and clean limits, respectively [22]. The slope of $H_{c2}(T)$ close to T_c is $dH_{c2}/dT = -1.1$ T/K and using $T_c = 4.6$ K, we found $\mu_0 H_{c2}(0) = 3.5$ T in the dirty and 3.7 T in the clean limit. The value of upper critical field is about half of the Pauli paramagnetic limit [23] of

Download English Version:

https://daneshyari.com/en/article/1817321

Download Persian Version:

https://daneshyari.com/article/1817321

Daneshyari.com