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# Superconductivity and ferromagnetism in Pd doped Y<sub>9</sub>Co<sub>7</sub>

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## Abstract

The ferromagnetic superconductor Y<sub>9</sub>Co<sub>7</sub> was chemically doped to yield the solid solution Y<sub>9</sub>Co<sub>7-x</sub>Pd<sub>x</sub> for 0 < x < 0.4. The lattice parameter *a* does not depend on x, whereas *c* increases with increasing Pd content up to x = 0.2, the palladium solubility limit. The transition from ferromagnetism (T<sub>C</sub> = 4.25 K) to superconductivity (T<sub>sc</sub> = 2.4 K) was observed only for the parent Y<sub>9</sub>Co<sub>7</sub> compound. For the lowest tested Pd doping level (x = 0.05), ferromagnetism is enhanced strongly (T<sub>C</sub> = 9.35 K) and superconductivity is not seen above 1.8 K. The Curie temperature rapidly increases from 4.25 K to about 10 K for a Pd concentration of x = 0.1 and remains almost unchanged for Y<sub>9</sub>Co<sub>6.8</sub>Pd<sub>0.2</sub>.

## Keywords:

A. intermetallics, B. magnetic properties, B. superconducting properties

## 1. Introduction

The intermetallic compound Y<sub>9</sub>Co<sub>7</sub>, despite its discovery 35 years ago [1], is still under investigation. This is due to its unusual properties, i.e., the coexistence of superconductivity (SC) and weak itinerant ferromagnetism (WFM) that is otherwise observed only for some 5*f* electron compounds [2],[3],[4],[5]. Bochenek, *et al.* [6] for example claim that Y<sub>9</sub>Co<sub>7</sub> is the

only known  $d$  band metal in which SC and WFM coexist on a microscopic scale.  $Y_9Co_7$ , previously known as  $Y_4Co_3$ , crystallizes in a hexagonal crystal structure (space group  $P6_3/m$ ) with lattice parameters  $a = 11.53 \text{ \AA}$  and  $c = 12.15 \text{ \AA}$  [7],[8]. The crystal structure of  $Y_9Co_7$  is presented in Fig. 1. It has a Curie temperature at about  $T_C \approx 4.25 \text{ K}$  and a superconducting transition temperature of about  $T_{sc} \approx 2 \text{ K}$  [1], and both strongly depend on the purity of the Y and Co starting materials and the post-annealing process [9].

There are many reports describing the crystal structure and the physical properties of  $Y_9Co_7$ , including its heat capacity, electrical conductivity, and magnetic susceptibility. Doping by Sc, Zr, Lu [10],[11],[12], Gd [11],[13] and La [11] on the Y site has been reported and the Co site has been partially doped by Si [7], Ni [11], [13], Mn, Fe, Cu, Zn and Al [11]. Yamaguchi, *et al.* have discussed the enhancement of ferromagnetism and the suppression of superconductivity as a function of the radius of the dopant [11]. They found that only the nonmagnetic lanthanides (La, Lu) and  $3d$  metals have a moderate influence the physical properties of  $Y_9Co_7$  while other metals dramatically increase the ferromagnetic  $T_C$  and decrease the superconducting  $T_{sc}$ .

Elemental palladium is an unusual metal that is located on the border of ferromagnetism. As an element it does not superconduct, but, because it typically contributes strongly to the electronic density of states at the Fermi energy, it is often a favorable constituent for enhancing superconductivity in ternary intermetallic compounds. For example, there are as many as 17 known palladium based superconductors in the Heusler family [14]. Herein, we report the synthesis, crystallographic and magnetic properties  $Y_9Co_{7-x}Pd_x$ . It is the first example of  $4d$  metal doping on the Co site in  $Y_9Co_7$ .

## 2. Experimental

A series of polycrystalline samples with composition  $Y_9Co_{7-x}Pd_x$  ( $x = 0, 0.05, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4$ ) was synthesized by arc-melting in a zirconium-gettered atmosphere of pure argon. The specimens were prepared using stoichiometric amounts of the pure elements (yttrium, cobalt, palladium). To homogenize the specimens, the obtained samples were remelted several times, then wrapped in tantalum foil, placed in evacuated quartz ampoules and annealed in the sequence  $620^\circ\text{C}$ ,  $520^\circ\text{C}$ ,  $420^\circ\text{C}$ , for a week at each temperature. The purity and crystallographic structure of the samples were tested by powder X-ray diffraction (Bruker D8 Focus,  $Cu K\alpha$  radiation, graphite-diffracted beam monochromator). Magnetic properties were tested by measuring ac magnetic susceptibility with a Quantum Design Magnetic Properties Measurement System (MPMS).

### 3. Results and discussion

The relationship between lattice parameters and the Pd concentration in  $Y_9Co_{7-x}Pd_x$  is presented in the Fig. 2. The lattice parameter  $a$  does not depend on  $x$  (Fig. 2(a)) within experimental resolution. The successful Pd doping is implied by modification of the  $c$  lattice constant, which changes linearly up to  $x = 0.2$  (Fig. 2(b)). It then saturates, indicating that a multiple phase thermodynamic region has been entered with higher Pd contents. Based on this observation,  $x = 0.2$  is the solubility limit of Pd in  $Y_9Co_7$  compound.

The temperature dependence of the real part of the  $ac$  magnetization ( $Re\_M$ ) for  $Y_9Co_{7-x}Pd_x$  ( $x = 0, 0.05, 0.1, 0.2$  and  $0.3$ ) is shown in Fig. 3. Measurements were limited to temperatures 2 K and higher, and in this temperature range a transition to the superconducting state is observed only for the parent  $Y_9Co_7$  with  $T_{sc} \sim 3$  K, confirming the good quality of our samples. For the same sample, the transition to the ferromagnetic state is seen by an increase of  $Re\_M(T)$  below 7 K, but the determination of the Curie temperature is not trivial; we estimate  $T_C$  by the minimum of the magnetisation derivative ( $d(Re\_M)/dT$ ) as shown in Fig. 3 for  $x = 0$  and  $0.2$ . The Curie temperatures are 5 K and 10.5 K for  $Y_9Co_7$  and  $Y_9Co_{6.8}Pd_{0.2}$ , respectively. A common method for determining the  $T_C$ s of itinerant ferromagnets is an Arrott plot [15], which is shown for the parent  $Y_9Co_7$  compound as the inset of Fig. 4. The Curie temperature obtained using this method ( $T_C = 4.25$  K) is only slightly lower than that estimated from the minimum of  $d(Re\_M)/dT$ , and proves correctness of the method used for a determination of  $T_C$ . For the samples containing Pd the PM-FM transition is sharp, whereas for the parent compound  $d(Re\_M)/dT$  is asymmetrical of which might be due to the presence superconducting fluctuations. The estimated Curie temperatures, plotted as a function of Pd content in  $Y_9Co_{7-x}Pd_x$ , are presented in Fig. 4. With increasing palladium content, up to the limit of solubility, the Curie temperature increases. For Pd contents greater than  $x = 0.2$ , the Curie temperature remains constant, a second indication that  $x = 0.2$  is the solubility of Pd in  $Y_9Co_{7-x}Pd_x$ .

In conclusion, we have synthesized and checked the crystal structure and magnetic properties of a series of samples with composition  $Y_9Co_{7-x}Pd_x$  (where  $0 < x < 0.4$ ). The  $a$  lattice parameter does not depend on  $x$ , whereas  $c$  increases linearly with increasing Pd content. This suggests that Pd atoms occupy sites in the electronically active Co(2b) chain and can therefore strongly influence physical properties of  $Y_9Co_7$ ; even  $x = 0.05$  results in the suppression of superconductivity to below 1.8 K. Moreover, with the increasing amount of dopant, the Curie temperature increases to over 10 K which is the highest  $T_C$  so far reported for doped  $Y_9Co_7$ .

If  $\text{Y}_9\text{Co}_7$  is a superconductor in proximity to ferromagnetism, then spin triplet superconductivity is expected, and very high sample quality is required for investigation of its properties due to the fact that impurities (both magnetic and non-magnetic) easily destroy superconductivity in such systems [16],[17]. Consistent with this picture, any kind of chemical doping suppresses the superconductivity and enhances the ferromagnetism of  $\text{Y}_9\text{Co}_7$ , but such information can only be considered indirect evidence for  $p$ -wave superconductivity in this system; to the best of our knowledge, direct experimental evidence for  $p$ -wave superconductivity in  $\text{Y}_9\text{Co}_7$  has never been reported. Given that Wiendlocha, *et al.* suggest that  $\text{Y}_9\text{Co}_7$  is a conventional, singlet superconductor, the problem remains open [18].

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Figures

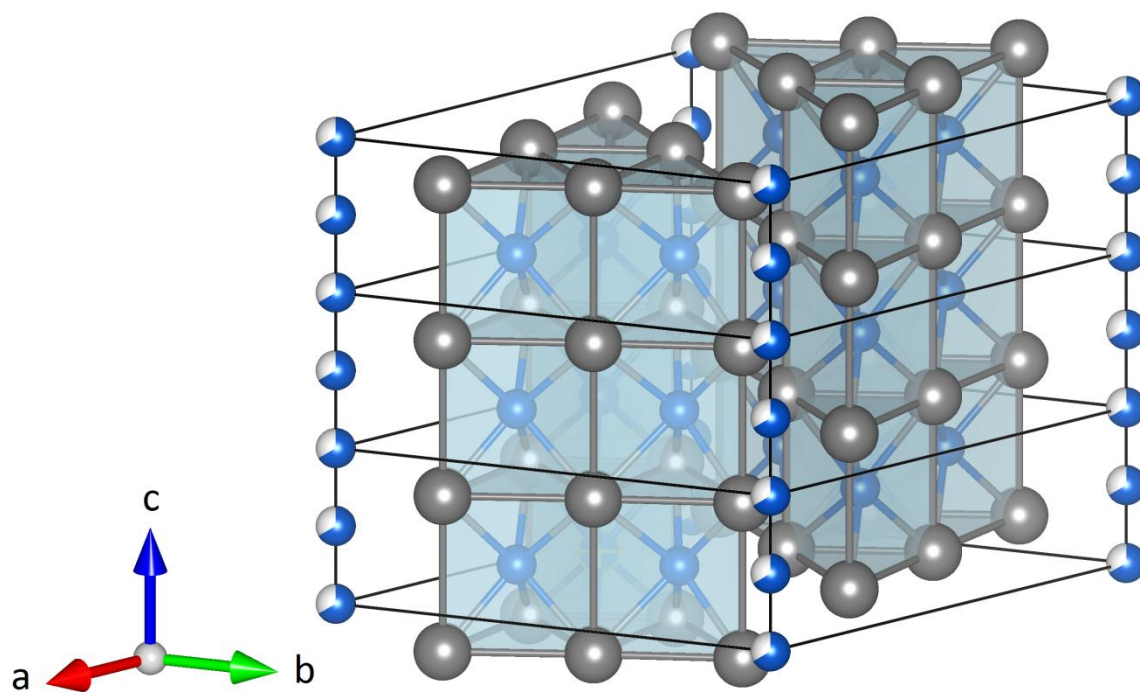


Fig. 1 Crystal structure of  $Y_9Co_7$ . Blue and grey balls represent cobalt and yttrium atoms, respectively.



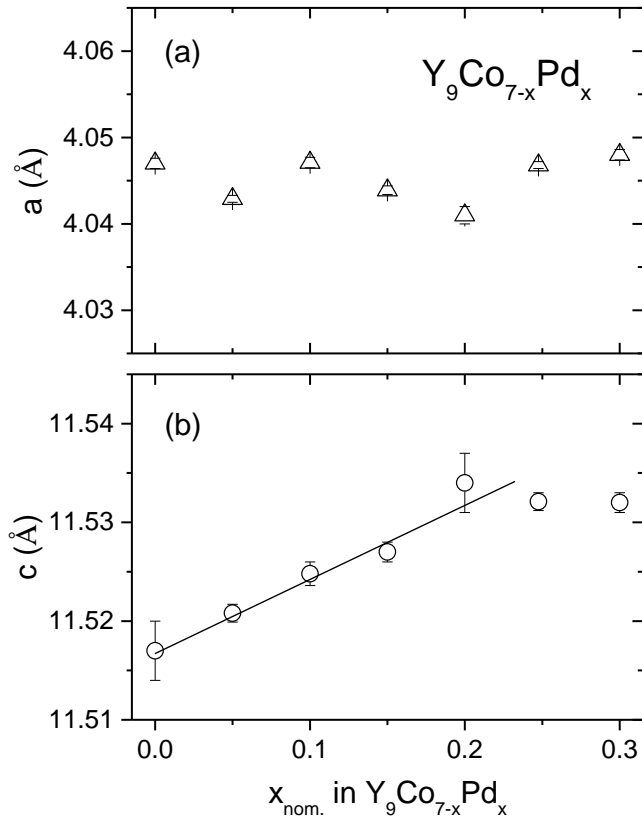


Fig. 2 The lattice parameters versus nominal palladium concentration ( $x$ ) in  $Y_9Co_{7-x}Pd_x$ .

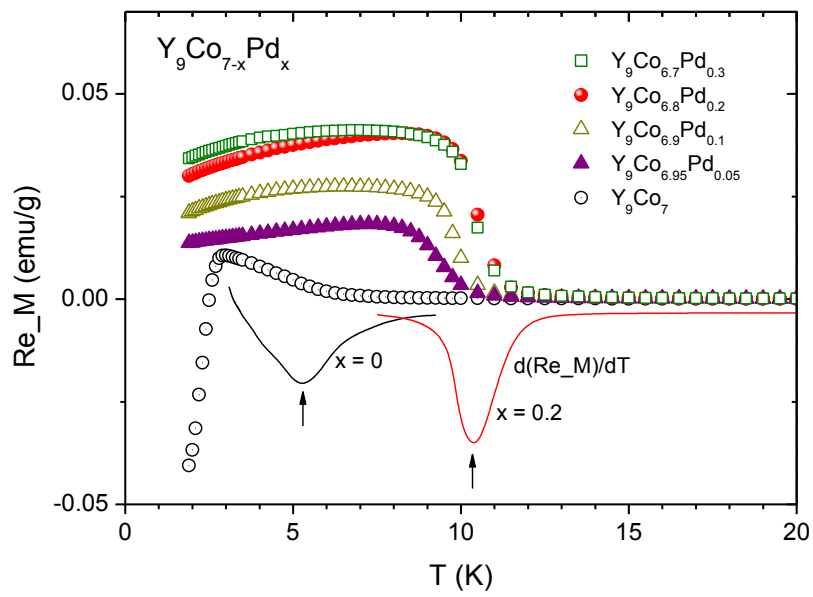


Fig. 3 Real part of the *ac* magnetization versus temperature (points) and temperature derivatives of the magnetization for selected Palladium content (solid lines).

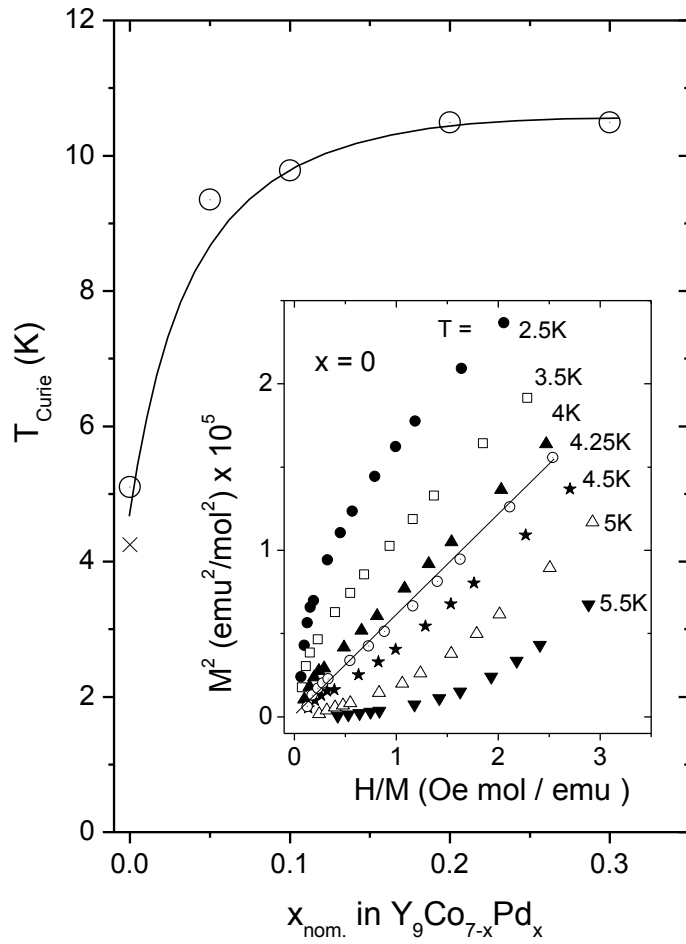


Fig. 4 The Curie temperature (open circles) versus nominal palladium concentration in  $\text{Y}_9\text{Co}_{7-x}\text{Pd}_x$ . Data points were estimated from the minimum of  $d(\text{Re}_M)/dT$ . The x shows the Curie temperature ( $T_C = 4.25\text{K}$ ) estimated from the Arrott plot presented in the inset for  $\text{Y}_9\text{Co}_7$ .

# Graphical Abstract

