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Doping effects on the magnetic properties of $NdRhIn_5$ intermetallic antiferromagnet

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ABSTRACT

We report temperature dependent heat capacity and magnetization measurements on single crystals of $Nd_{1-x}La_xRhIn_5$ ($x = 0.15$, 0.4 and 0.5) and $NdRhIn_{5-x}Sn_x$ ($x = 0.08$, 0.12 and 0.24). $NdRhIn_5$ is an antiferromagnetic (AFM) compound with $T_N \approx 11$ K which crystallizes in the same layered tetragonal structure of the $CeMIn_5$ family (M = Rh, Co and Ir), where different ground states can be found by tuning the interplay among different microscopic interactions such as the Kondo effect, crystal field (CEF) effects and the Ruderman–Kittel–Kasuya–Yoshida (RKKY) magnetic interaction. Here, we explore the evolution of the AFM correlations in this Nd-based (non-Kondo) compound while perturbing the RKKY exchange by using two different substitutions: (i) replacing Nd^{3+} by non-magnetic La^{3+} within $NdIn_3$ atomic planes (dilution) and (ii) substituting In by Sn in the In-sites (electronic tuning). For both types of doping, our results show the suppression of the AFM state as the La- or Sn-content is increased. This doping induced suppression of the AFM order is discussed considering the effects of dilution and effects in the tetragonal CEF using a mean-field model applied to the observed data. Our results are compared to the properties of other members of the $RRhIn_5$ family considering the role of dimensionality in the magnetic interactions.

1. Introduction

Dilution in magnetically ordered system is one of the important subjects within the study of percolation phenomena. It represents a way to understand the nature of long and short range antiferromagnetic (AFM) ordering in strongly correlated electrons systems, where it might be used to suppress the Néel state and probe interesting different ground states near to the AFM quantum critical point (QCP). In this regard, a series intensively investigated in the last eight years is the family of heavy-fermion superconductors $CeMIn_5$ (M = Rh, Co, Ir), where magnetic dilution by chemical doping has proved to be a valuable tool as tuning parameter in the interplay between the Kondo effect and the long range Ruderman–Kittel–Kasuya–Yoshida (RKKY) magnetic interaction. The balance between these two interactions as a function of doping may tune the ground state of a system from heavy-fermion paramagnetic metal or unconventional superconductors to a long range ordered AFM state [1–10].

Recently, La- and Sn-doping studies on $CeRhIn_5$ under pressure were reported [11]. These results performed on single crystals of $Ce_{0.9}La_{0.1}RhIn_5$ and $CeRhIn_{4.84}Sn_{0.16}$ have shown that although both compounds have the same $T_N = 2.8$ K, Sn-doping shifts the pressure induced superconducting phase to lower pressures (compared to those required in the undoped $CeRhIn_5$) while La-doping does exactly the opposite, shifting the $P-T$ phase diagram to higher pressure. These observations reveal that the strength of the Kondo coupling is the relevant energy scale to set the pressure range for the occurrence of superconductivity (SC) in $CeRhIn_5$ [11]. These results indicate that one important step to understand the doped-induced evolution of the physical properties of the Ce-based $RMIn_5$ family may be the analysis of dilution effects on the magnetic interaction between rare earth ions in non-Kondo isostructural materials from the $R_mM_nIn_{3m+2n}$ (R = Ce−La; M = Co, Rh or Ir; $m = 1, 2$; $n = 0, 1$) family, where the absence of strong hybridization between the localized 4f electrons and the conduction 5d electrons make them a reference compounds to study the suppression of antiferromagnetism in these series. Our recent work on $Tb_{1-x}La_xRhIn_5$ ($0.0 < x < 1.0$) [12], where we evaluated the role of the different mechanisms for the suppression of the long-range AFM coupling by considering dilution, changes in the CEF scheme and the introduction of disorder is a relevant example on the subject. In $Tb_{1-x}La_xRhIn_5$ series, the Néel temperature decreases with a non-linear behavior as a function of La-concentration and extrapolates to zero at

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roughly 70% of La content (the site percolation threshold for a 3D magnetic system), differently to the $x_c \sim 40\%$ observed for $Ce_{1-x}La_xRhIn_5$ and $(Ce_{1-x}La_x)_2RhIn_8$ families. X-ray magnetic scattering experiments for the determination of magnetic structure on a sample with 40% of La-content revealed that the magnetic wave vector is the same as for the undoped compound. Also, it was conclude that the crystal field scheme evolves as a function of La-doping and affects T_N as much as the decreasing in J_{RKKY} due to dilution [12]. In $NdRhIn_5$ compound, which is also an isostructural non-Kondo member of the $CeMIn_5$ heavy-fermion series and orders antiferromagnetically below $T_N \sim 11$ K, Nd^{3+} ($J = \frac{9}{2}$) is a Krammer ion such as Ce^{3+} ($J = \frac{5}{2}$) and the CEF effects act in distinct way than that for Tb^{3+} ($J = 6$) which is a non-Krammer ion. As such, dilution studies on $NdRhIn_5$ may represent a interesting complementary study to the dilution studies on $TbRhIn_5$ [12]. In this work, we have explored the doping effects on the magnetic interactions between Nd^{3+} ions by two different chemical substitutions: replacing Nd^{3+} by non-magnetic La^{3+} ions for $0 \leq x \leq 0.5$, and substituting In by Sn for $x = 0$, 0.08, 0.12, 0.24 and 0.48. The doping evolution of the magnetic properties of $NdRhIn_5$ was studied by measuring magnetic susceptibility and specific heat as a function of temperature. The reported data was interpreted considering the doping effects on the long-range AFM RKKY interaction and on the CEF scheme as a function of dopant concentration in both series.

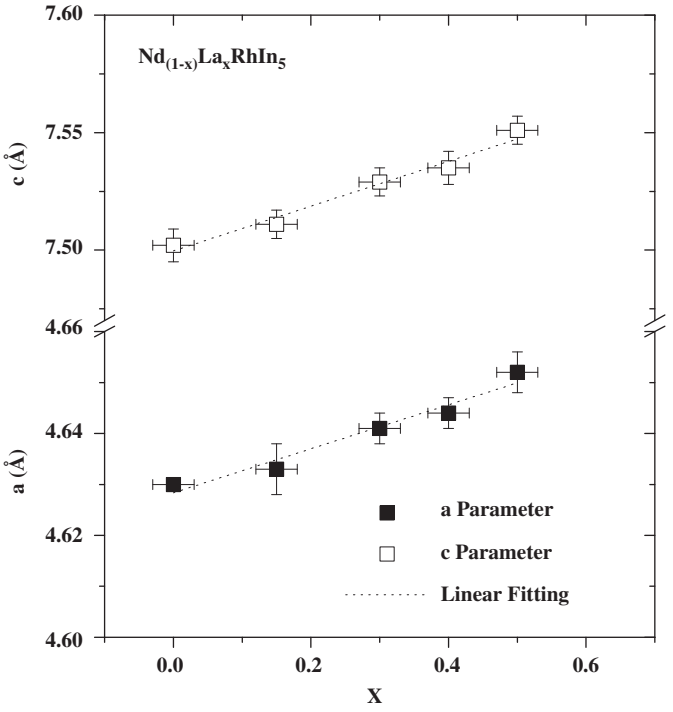

Fig. 1. Cell parameters of $Nd_{1-x}La_xRhIn_5$ ($x = 0$, 0.15, 0.3, 0.4, 0.5) as a function of x.

2. Experimental results and discussion

Single crystals of $Nd_{1-x}La_xRhIn_5$ ($x = 0$, 0.15, 0.3, 0.4 and 0.5) and $NdRhIn_{5-x}Sn_x$ ($x = 0$, 0.08, 0.12, 0.24 and 0.48) were grown by the self-flux method [13]. In the case of the $Nd_{1-x}La_xRhIn_5$ series, x denotes the nominal concentration of Lanthanum. For $NdRhIn_{5-x}Sn_x$ compounds, the starting materials were mixtured in the ratio Nd: Rh: In: Sn = 1: 1: 20y, and x was taken as $x \sim 0.4y$ as in the case of $CeRhIn_5$ [7]. The grown single crystals were confirmed by powder X-ray diffraction to crystallize in the $HoCoGa_5$-type structure with no traces of the Nd_2RhIn_8 or $NdIn_3$ phases. For the magnetic characterization of both systems, magnetization measurements were performed in a commercial SQUID magnetometer at $H = 1$ kOe in the temperature range between 2 and 300 K. Specific heat measurements were performed in a quantum design PPMS small-mass calorimeter that employs a quasi-adiabatic thermal relaxation technique; C/T data were taken between $1.8 \leq T \leq 40$ K.

The first observed effect from the introduction of the larger La^{3+} ion in the $NdRhIn_5$ structure is the linear increase of the tetragonal cell parameters, a and c (Fig. 1), in agreement with the Vegard's law for solid solutions. a and c were determined from least-squares minimization of the Bragg peak positions (2θ) [14]. For $NdRhIn_{5-x}Sn_x$, our X-ray powder diffraction patterns and cell parameters calculations revealed that there are no appreciable changes in the lattice parameters within the Sn-concentration range. Horizontal error bars in Fig. 1 has been estimated from linear fits to the inverse of the magnetic susceptibility for $T > 200$ K and assuming the full calculated moment of $3.66\mu_B$ for the free Nd^{3+} ion. Vertical error bars are extracted from the cell parameters calculations.

In Fig. 2 we show the results of the macroscopic measurements (magnetic susceptibility and specific heat) of three representative samples from each studied family. The three panels to the left contain the data of the $Nd_{1-x}La_xRhIn_5$ ($x = 0$, 0.15 and 0.4) series while the ones to the right show the data for $NdRhIn_{5-x}Sn_x$ ($x = 0$, 0.12 and 0.24) samples. Data for $NdRhIn_5$ taken from Ref. [15] were also included. For all cases, the main panels depict the data of magnetic susceptibilities for $H = 1$ kOe applied parallel ($\chi_{\parallel}$) and perpendicular ($\chi_{\perp}$) to the c-axis, while the insets contain the specific heat data. The solid curves represent the best fits to the experimental data using the mean field (MF) model of Ref. [16] which includes a mean field first-neighbors exchange interaction and the tetragonal CEF Hamiltonian. The temperature of the maximum in both $\chi(T)$ and C/T curves coincides well and we have taken these temperature values as the Néel temperature T_N.

From our $\chi(T)$ and C/T, becomes evident the shift of T_N to lower-T as the doping content is increased, indicating the weakening of long range magnetic correlations between Nd^{3+} ions as La or Sn atoms enter the structure. The broadening of the transition in the T_N data is more evident for the La-doped samples than for the Sn-doped ones, which is consistent with a smaller dilution limit in the former compounds (see below). Interesting, the MF simulations in both cases follow the details of the experimental data quite well, which is indicative of the evolution of CEF effects as a function of doping.

In Fig. 3 we present the x-dependence of the normalized Néel temperature ($T_{N,x}/T_{N,x=0}$) as extracted from the data of Fig. 2 for all the La-doped (filled circles) and Sn-doped (open circles) studied samples. Additionally, we show the reported T_N values from the series $CeRhIn_{5-x}Sn_x$ [7] (open diamonds), $Ce_{1-x}La_xRhIn_5$ [4] (filled diamonds) and $Tb_{1-x}La_xRhIn_5$ [12] (stars) for comparison. For both Ce-based families, T_N shows a linear decrease (dashed lines) as a function of x. This is also the case for the $NdRhIn_{5-x}Sn_x$ compounds, while a non-linear (the dashed-dotted curves) T_N suppression as a function of x is found in $Nd_{1-x}La_xRhIn_5$ and $Tb_{1-x}La_xRhIn_5$. For the $Nd_{1-x}La_xRhIn_5$ series, we observed no long range order for $x = 0.5$, thus the critical concentration at which $T_N \to 0$ must be between $0.4 < x_c < 0.5$, roughly the same x_c found in the $Ce_{1-x}La_xRhIn_5$ ($x_c \sim 0.4$) [4]. However, above $x = 0.3$, T_N suddenly drops to zero with a non-linear behavior, contrasting with the linear decrease observed in Ce-based materials. For $Tb_{1-x}La_xRhIn_5$, the same evolution has been observed with a higher dilution limit of $0.7 < x_c < 0.8$ [12]. For $NdRhIn_{5-x}Sn_x$ compounds, on the other hand, we could not determine, in this work, the dilution limit up to the higher studied concentration