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Structure and physical properties of RE_2AgGe_3 (RE=Ce, Pr, Nd) compounds

Sumanta Sarkar [a], Dundappa Mumbaraddi [a], Pramod Halappa [a], Deepti Kalsi [a], Sudhindra Rayaprol [b], Sebastian C. Peter [a,*]

[a] New Chemistry Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Jakkur, Bangalore 560064, India
[b] UGC-DAE Consortium for Scientific Research, Mumbai Centre, R-5 Shed, BARC Campus, Mumbai 400085, India

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ABSTRACT

We have synthesized the compounds RE_2AgGe_3 (*RE* =Ce, Pr, Nd) by arc melting. The crystal structure obtained from single crystal and powder X-ray diffraction suggests that these compounds crystallize in the α-$ThSi_2$ structure type. The magnetic susceptibility data of Ce_2AgGe_3 follows Curie–Weiss (CW) law above 25 K without any magnetic ordering down to 2 K. The effective magnetic moment (μ_{eff}) was calculated as 2.53 μ_B/Ce and negative Curie paramagnetic temperature (θ_p)= −2.4 K hint weak antiferromagnetic coupling among the adjacent spins. Pr_2AgGe_3 shows a complex magnetic behavior wherein the magnetic susceptibility at field cooled and zero field cooled modes bifurcates at 11.5 K with the latter undergoing a cusp like maxima, probably due to weak ferromagnetic interaction. The θ_p and μ_{eff} obtained are 4 K and 4.33 μ_B/Pr, respectively. Nd_2AgGe_3 undergoes multiple magnetic transitions. Temperature dependent resistivity data reveals that three compounds are metallic in nature.

1. Introduction

Rare-earth based compounds are one of the building blocks of material science due to their advanced application in the field of superconducting materials, magnetic materials, luminescent materials and hydrogen storage [1]. Recently, rare-earth based intermetallic compounds have been attracting much attention because of their versatile structural and physical properties such as intermediate valency [2], heavy fermion, Kondo behavior [3,4], spin glass behavior [5], superconductivity [6], structural phase transition [7], giant magnetoresistance [8], magnetocaloric effect [9] and zero thermal expansion [10]. These properties are associated with the presence of localized electronic 4*f* shell interaction with itinerant *s*, *p*, *d*-conduction electrons. Variable valences of cerium ($Ce^{3+/4+}$) can lead to many peculiar properties [11].

In this regard, it is interesting to briefly discuss about compounds crystallizing in the $ThSi_2$ structure type (both α and β-phases), particularly those with the general formula, RE_2TX_3 (RE=Ce, Pr, Nd; T=Transition metal, X=Si, In, Ge) owing to their fascinating structural and physical aspects. Here we give a brief overview on the compounds based on Ce, Nd and Pr. The examples are atom-disorder spin glass behavior was observed in Ce_2CuSi_3, Pr_2CuSi_3, Nd_2CuSi_3; Kondo lattice compound Ce_2NiGe_3 showed spin glass behavior; short range antiferromagnetic ordering was observed in Nd_2NiGe_3; annealed sample of Ce_2NiSi_3 showed antiferromagnetic transition at 4.2 K and additional anomaly at 2.5 K, Ce_2FeSi_3 and Ce_2PdSi_3 exhibit Kondo behavior and ferromagnetic ordering was observed below 16 K in Nd_2PdSi_3 [12–19], Kondo lattice reported in Ce_2IrSi_3 and Ce_2CoSi_3 [20,21]; weak spin glass behavior was observed in Nd_2PtSi_3 [22]. Ce_2RhSi_3 is antiferromagnetic below $T_N < 7$ K, ferromagnetic spiral magnetic ordering was observed at 4.2 K observed in a Nd_2RhSi_3 [21,23] and Ce_2CuGe_3 was reported as to show spin glass behavior [24]. All these compounds crystallize in the AlB_2 type structure (β-$ThSi_2$). Compounds crystallizing in the α-$ThSi_2$-type structure are $CeFe_{0.22}Si_{1.78}$ [25], $CeCu_{0.14}Ge_{1.71}$ [26], $CeTi_{0.23}Ge_{1.77}$ [27], $CeRh_{0.5}Ge_{1.5}$ [28], $PrNi_{0.25}Si_{1.75}$ [29], $PrNi_{0.07}Ge_{1.79}$ [30], $PrAg_{0.24}Ge_{1.76}$ [31], $PrCo_{0.3}Ge_{1.7}$ [32], $NdZn_{0.3}Si_{1.7}$ [33], $NdZn_{0.45}Si_{1.55}$ [34], $NdNi_{0.25}Si_{1.75}$ [35] and $NdNi_{0.25}Si_{1.75}$ [29]. Among the α-$ThSi_2$-type compounds, we have reported the spin glass behavior in $CeRh_{0.5}Ge_{1.5}$ [28]. The above mentioned works suggest that there may be other interesting compounds in the α-$ThSi_2$-type structure with interesting physical properties.

In comparison to the compounds with first row transition metals (e.g. Mn, Fe, Co, Ni, Cu), 4*d* and 5*d* block elements (e.g. Ru, Rh, Pd, Ag, Ir, Pt, Au) present an intriguing aspect to the overall physical properties of the compound because of the fact that 4*d*

* Corresponding author. Fax: +91 80 22082627.
E-mail address: sebastiancp@jncasr.ac.in (S.C. Peter).

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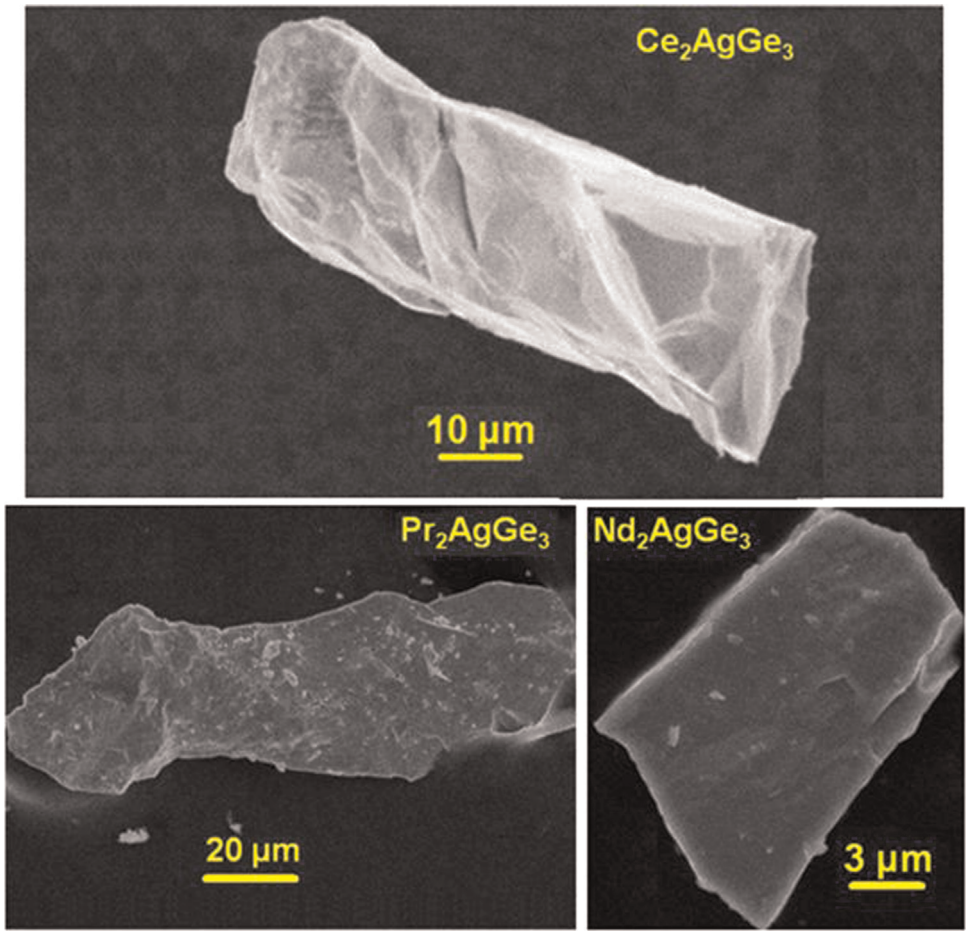

Fig. 1. FE SEM images of the single crystals of Ce_2AgGe_3, Nd_2AgGe_3 and Pr_2AgGe_3.

and 5*d* electrons can efficiently hybridize with the localized 4*f* orbitals belonging to lanthanide metals. This type of interaction dictates the conductivity and related properties of the itinerant electrons giving rise to interesting and very often anomalous properties. The other motivation of this work was our continuous search for new superstructures of the AlB_2 type structure similar to our recent studies on ordered compounds Eu_2AuSi_3, Eu_2AgGe_3, Eu_2AuGe_3, Yb_2AuSi_3 and Yb_2AuGe_3 [36–38], even though they were reported in the disordered structures earlier [29,39–41]. We also observed the lack of ordered structure as in $CeAu_xGe_{1-x}$ [42], $CeRh_xGe_{1-x}$ [28] and Nd_2NiGe_3 [43]; the first two led to the formation of the mixture of hexagonal and tetragonal crystal systems, but later one crystallize only with AlB_2 type. Understanding all these analyses, we have synthesized three Ag based compounds RE_2AgGe_3 (RE=Ce, Pr, Nd) by arc melting method. Our detailed powder and single crystal XRD measurements suggest that they are crystallizing in the α-$ThSi_2$ structure type. All three compounds found to be pure and further studied for their magnetic and transport properties in detail. All three compounds show diverse magnetic properties and are metallic in nature.

2. Experimental

2.1. Synthesis

The following metals were used as purchased without any further purification; Rare earth (Ce, Pr and Nd chunks, 99.99%, Alfa-Aesar), Ag (shots, 99.99%, Alfa-Aesar) and Ge (pieces, 99.999%, Alfa-Aesar). In a typical procedure, rare earth, silver and germanium metals were taken in an ideal 2:1:3 atomic ratios (total weight of reactant was ~200 mg) and repeatedly arc melted (flipping 5 times with 30 s arc passing) in an argon atmosphere to ensure homogeneity; hard globules were formed. Finally, the samples were crushed and made into powder for further characterization. All the samples are stable under normal atmospheric conditions for several months.

2.2. Elemental analysis

Semi quantitative microanalyses were performed on Ce_2AgGe_3, Pr_2AgGe_3 and Nd_2AgGe_3 single crystals using a Leica 220i electron microscope (SEM) equipped with Bruker 129 eV energy dispersive X-ray analyzer (EDS). Data were acquired with an accelerating voltage of 20 kV and 90 s accumulation time. The EDS analysis performed on cleaned surfaces of the single crystals percent showed the atomic composition for Ce_2AgGe_3 is 33.16(2) Ce, 20.90 (1) Ag and 45.94(1) Ge; for Pr_2AgGe_3 31.64(2), 19.17(1) and 49.19 (1); for Nd_2AgGe_3 33.62(3) Nd, 18.20(1) Ag, 48.18(1) Ge. The compositions obtained from EDS data are in good agreement with the results derived from the refinement of single crystal X-ray diffraction data. Field emission scanning electron microscopy (FE SEM) images of the representative single crystals are shown in Fig. 1.

2.3. Powder X-ray diffraction

The phase identity and purity of the samples were confirmed by powder XRD measurements carried out with on a Bruker D8 Discover diffractometer using Cu-Kα radiation (λ=1.5406 Å). All