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Interplay of Sm^{4f} and Co^{3d} spins in SmCoAsO

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ABSTRACT

We present detailed magnetization and magneto-transport studies on the title compound SmCoAsO. In a recent paper we reported (Awana et al., 2010 [1]) the complex magnetism of this compound. SmCoAsO undergoes successive paramagnetic (PM)–ferro-magnetic (*FM*)–anti-ferro-magnetic (*AFM*) transitions with decrease in temperature. This is mainly driven via the *c*-direction interaction of Sm^{4f} (SmO layer) spins with adjacent (CoAs layer) ordered Co^{3d} spins. In this article we present an evidence of kinetic arrest for *FM–AFM* transition. The isothermal magnetization (*MH*) loops for SmCoAsO exhibited the meta-magnetic transitions at 6, 8 and 10 K at around 80, 60 and 50 kOe fields, respectively, with characteristic hysteresis shoulders along with the non-zero moment at the origin, thus suggesting the possibility of kinetic arrest. Suggested kinetic arrest is further evident in zero field-cooled (*ZFC*) and field-cooled (*FC*) hysteresis under high fields of up to 140 kOe magnetization (*MT*) and the magneto-transport measurements *R*(*T*)*H* during *FM–AFM* transition. The time dependent moment experiments exhibited very small (~2–3%) increase of the same below 20 kOe and decrease for 30 kOe at 15 K.

1. Introduction

Since the recent discovery [2] of superconductivity in doped REFeAsO (RE=rare earths) oxy-pnictides, similar structure cobaltates (RECoAsO) have attracted renewed interest [3–5]. Co orders ferro-magnetically (*FM*) below 100 K in case of LaCoAsO with a saturation moment of 0.10–0.20μ_BB below Curie temperature (T_c) and relatively higher effective paramagnetic moment of above 1.50μ_B/Co above T_c, thus indicating an itinerant *FM* state [1,3–5]. The situation becomes more interesting and rather complex with gradual *FM–AFM* transitions, when the non-magnetic La is replaced with magnetic Nd [6,7], Sm [1,5,8] or Ce [5,9]. It seems that magnetic RE spins influence the *FM* ordering of Co sub-lattice and convert the same to *AFM* at lower temperatures. In such situations a natural question arises as if the *FM–AFM* state is kinetically arrested or not. This has been evidenced in a number of well experimented magnetization studies on say $CeFe_2$ [10,11] and some magnetically phase separated manganites [12], where an *FM–AFM* transformation takes place gradually at low temperature. In this article we address the kinetic arrest of *FM–AFM* transition in SmCoAsO and compliment our earlier recent work on the same compound [1]. Our high field magnetization and magneto-transport studies establish possible FM–AFM kinetic arrest in SmCoAsO for the first time.

2. Experimental

Polycrystalline SmCoAsO is synthesized by the single step solid-state reaction method via the vacuum encapsulation technique [1]. Stoichiometric amounts of high purity (~99.9%) Sm, As, Co_3O_4 and Co powders are ground thoroughly using mortar and pestle. The mixed powders were palletized and vacuum-sealed (10^{-4} Torr) in a quartz tube. This sealed quartz ampoule was placed in a box furnace and heat treated at 550 °C for 12 h, 850 °C for 12 h and then finally 1150 °C for 33 h in continuum with slow heating rate; details of synthesis are further given in Ref. [1]. The entire heating process is "Single Step" in nature as reported earlier by some of us [13]. The X-ray diffraction pattern of the compound was taken on a Rigaku X-ray diffractometer with Cu K_α radiation. The resistivity measurements were carried out by a conventional four-probe method on Physical Property Measurement System (*PPMS*) from Quantum Design—USA. Heat capacity and magnetization measurements were also carried out on the same *PPMS*.

3. Results and discussion

As-synthesized SmCoAsO is crystallized near single phase with lattice parameters $a=3.957(3)$ Å and $c=8.242(2)$ Å [1]. Magnetic moments (*M*) versus temperature (*T*) plots for SmCoAsO in field-cooled situation at varying applied fields from 10 Oe to 90 kOe are depicted in Fig. 1. The compound undergoes *PM*–FM–*AFM*

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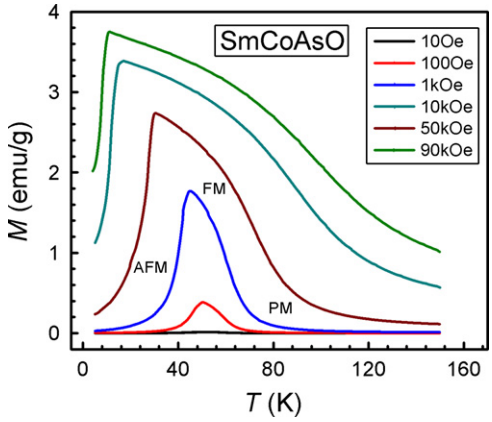

Fig. 1. $M(T)$ down to 5 K in field-cooled situation at fields from 10 Oe to 90 kOe for SmCoAsO.

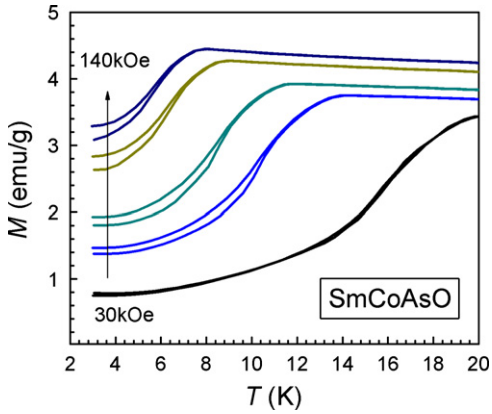

Fig. 2. $M(T)$ in both *ZFC* and *FC* situations at 30, 80, 100, 120 and 140 kOe fields for SmCoAsO.

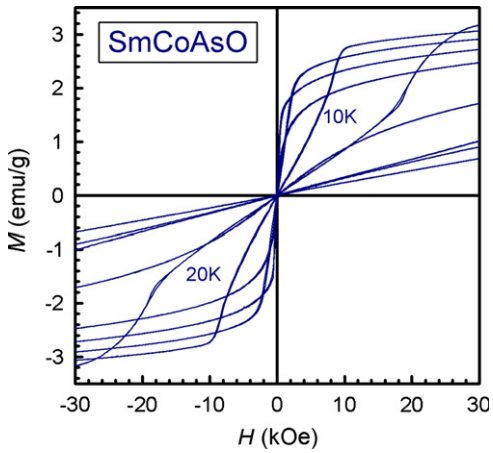

Fig. 3. $M(H)$ for SmCoAsO till 30 kOe applied fields in all five quadrants, meta-magnetic field is seen clearly at 20 K and around 20 kOe with hysteresis in shoulder as marked.

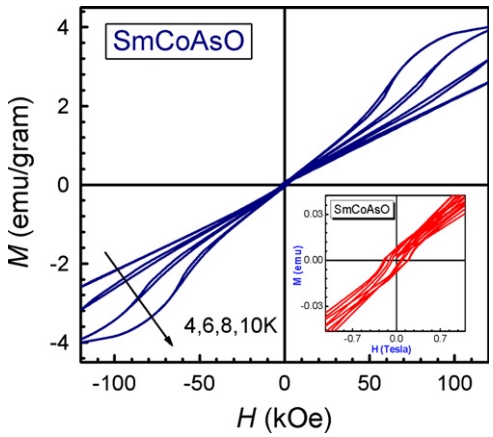

Fig. 4. $M(H)$ for SmCoAsO till 120 kOe applied field in all five quadrants, the meta-magnetic transitions are seen clearly at 6, 8 and 10 K at around 80, 60 and 50 kOe fields, respectively, with characteristic shoulders and hysteresis.

transitions with lowering of temperature. At low fields of 10 Oe it exhibits two peaks at around 57 and 45 K [1]. For higher fields of above 100 Oe only one single peak for *FM–AFM* transition is evident. With increase in applied field the *FM–AFM* transition peak temperature decreases below 10 K for 100 kOe field. This is in accordance with earlier reports on similar compounds [1,5,8].

To check if the *FM* is kinetically arrested during the *FM–AFM* transition, we carried out the magnetization measurements in both field-cooled (*FC*) and zero-field-cooled (*ZFC*) situation in higher fields of up to 140 kOe. These results are depicted in Fig. 2. For an applied field of 30 kOe there is nearly no branching of *FC* and *ZFC*. However as the field is increased to 60, 80, 120 and 140 kOe the *ZFC* and *FC* branching is clearly evident at 11, 9, 7 and 6 K, respectively. The *FC* and *ZFC* branching seen at 140 kOe field cannot be due to spin-glass (SG) magnetic phase [11,12]. At this juncture we tend to believe that possibly the *FM* phase is kinetically arrested during the *FM–AFM* transition in SmCoAsO. It is also evident from Fig. 2 that the kinetic arrest of *FM* phase if at all present is for higher fields, i.e. >30 kOe. To further establish the possible kinetic arrest, we cooled the sample under 140 kOe field from 250 K, i.e. from within paramagnetic state and carried out the *MT* down to 3 K and the isothermal magnetization (*MH*) is recorded from the cooling field 140 kOe to zero and checked if there is any non-zero moment left at the origin. We found that the compound bears a non-zero moment at origin. The high field isothermal magnetization (*MH*) results for SmCoAsO will be discussed later.

Relatively lower field (<30 kOe) isothermal magnetization (*MH*) results for SmCoAsO are presented in Fig. 3 for various temperatures in *PM*, *FM* and *AFM* regions. The *MH* is linear in both paramagnetic (150 K) and anti-ferromagnetic (2 and 5 K) regions. For *FM* regions (40, 60 and 80 K) it is the same with saturation moment of $\sim 0.20\ \mu_B$. This is similar to that as reported earlier for similar compounds [6–9]. The *MH* plots exhibit clear meta-magnetic transitions at 10 and 20 K, respectively, at 10 and 20 kOe fields. These plots are marked in Fig. 3. Also the *MH* plots at 20 K exhibit characteristic shoulder hysteresis at 20 kOe field. Interestingly these temperatures of 10 and 20 K fall in the region where the compound is in the middle of the *FM–AFM* transformation. Clearly in this region the competing *FM* and *AFM* states give rise to meta-magnetic instability, which is being reminiscent in the *MH* plots in Fig. 3.

High field (120 kOe) field isothermal magnetization (*MH*) plots for SmCoAsO at 4, 6, 8 and 10 K are depicted in Fig. 4. This is precisely to check possible reminiscence of *FM* clustering within *AFM* dominated phase or the kinetic arrest of *FM*. The *MH* plot at 4 K is nearly linear without any meta-magnetic deviation or visible hysteresis. The *MH* plot at 6 K is also though nearly linear but with hysteresis opening between 40 to 120 kOe. Both the 8 and 10 K *MH* plots exhibit shallow/rounded meta-magnetic like steps at around 80 and 60 kOe, respectively, with hysteresis at these shoulders. These hysteresis shoulders are similar to that as observed at 20 K for low field (<30 kOe) *MH* data shown in Fig. 3. The meta-magnetic