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Magnetic properties of RTSb₃

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Magnetization, electrical resistivity, and thermal expansion measurements have been performed on polycrystalline RTSb₃ (R=La, Ce, Pr, Nd, Gd, Tb, and Dy; T=Cr, V) samples in order to study their magnetic properties. Depending on the rare-earth element, RTSb₃ has been found to have either a purely ferromagnetic (or antiferromagnetic) phase or combination of antiferromagnetic (lower-temperature) and ferromagnetic (higher-temperature) phases. The antiferromagnetic order evolves from the ferromagnetic order as a result of the competition between R³⁺ and Cr³⁺ ions. As R is changed from La to Dy, the antiferromagnetic ordering of the R³⁺ ions becomes more prominent and predominate over the ferromagnetic ordering of Cr³⁺ for R=Gd, Tb, and Dy.

Thermal expansion measurements show that the antiferromagnetic transition accompanies a drop in the sample dimension. The order of the ferromagnetic transition is found to be of the second order.

INTRODUCTION

The intermetallic alloys formed by combining the magnetic properties of the 4f metals (large magnetocrystalline anisotropy, R) and the 3d metals (large exchange coupling, T) in RTSb₃ present an interesting class of magnetic material both for the study of their basic magnetic behavior and for their possible applications. It is mentioned that the rare-earth metal is useful in stabilizing low-dimension features such as layers or chains (seen in CeCrSb₃). Successive layering of such compound allows for the control of the magnetic aspect through interlayer coupling.

Previous results on the magnetic properties of the RTSb₃ system with T=Cr and V show that the Cr atoms which order ferromagnetically, correspond to two unpaired spins, whereas the V has one uncompensated spin. The lowered Curie temperatures for RTSb₃ (T=Cr) are attributed to the R–T coupling where R orders antiferromagnetically. In the same way as the atomic radius decreases from La to Lu, it was found that substitution of heavier rare earths for La in LaT₃Sb₃ causes the structure of the compound to contract. The contraction of the structure is accompanied by changes in the magnetic properties of the system. This led us to extend the study of the magnetic properties of RTSb₃ with R=La, Ce, Pr, Nd, Gd, Tb, and Dy.

EXPERIMENTAL DETAILS

Polycrystalline RTSb₃ samples were synthesized by arc melting the proper amounts of the constituent elements in an Ar atmosphere. An excess amount (10%) of Sb was added to compensate for its weight loss during melting. The purities of the elements were 99.9% for all the rare-earth elements, 99.99% for Cr, 99.9% for V, and 99.999% for Sb. Samples were annealed in an evacuated quartz tube at 600 °C for two days and at 800 °C for eight days. Magnetization measurements were carried out by means of a superconducting quantum interference device magnetometer (Quantum Design, CA) over the temperature range of 5–400 K and in an applied magnetic field up to 5.5 T. Thermal expansion measurements were made using a capacitance dilatometer over the temperature range of 4.2–200 K. Electrical resistivity measurements were done using the standard four-probe method over the temperature range of 5–300 K.

RESULTS AND DISCUSSIONS

Figure 1 shows the magnetization behavior of the RCrSb₃ (with R=La, Ce, Pr, Nd, Gd, and Tb) compounds as a function of temperature. Unlike specified, all samples were cooled in zero applied magnetic field from room temperature and magnetization measurements were done in an applied field of 0.01 T while raising the temperature. Figure 1(a) shows that LaCrSb₃ is ferromagnetic with a Curie temperature of 144 K. The transition temperature was determined from the first derivative of M vs T data. The Tc found by Hartjes et al. for LaCrSb₃ is 125 K, which was determined from the lowest point of the 1/Θ vs T plot. In Fig. 1(b) CeCrSb₃ shows ferromagnetic ordering with Tc=140 K. This is much higher than that found by Hartjes et al. The inset shows the magnetization data of CeCrSb₃ at 1 T. It shows that there is another ordering below 30 K. Interestingly, the PrCrSb₃ sample [Fig. 1(c)] shows antiferromagnetism below 16 K while it is ferromagnetic at higher temperatures with Tc=142 K. A comparison of the low-temperature ordering in CeCrSb₃ [the inset of Fig. 1(b)] with the low-temperature antiferromagnetic ordering in PrCrSb₃ suggests that in CeCrSb₃ there is an onset of an antiferromagnetic phase at low temperature. Temperature-dependent magnetization of the NdCrSb₃ [Fig. 1(d)] sample is similar to that of PrCrSb₃. For NdCrSb₃, transition temperatures of T_N=18 K and T_c=141 K were found. The overall magnetic behavior of the samples presented in Figs. 1(a), 1(b), 1(c), and 1(d) agree with those found by Hartjes et al.

We now turn our attention to new systems in the RTSb₃ series, namely, GdCrSb₃, TbCrSb₃, DyCrSb₃, and DyVSb₃.
In Fig. 1(e), GdCrSb$_3$ shows an antiferromagnetic ordering with $T_N=26$ K. GdCr$_3$ does not show the ferromagnetic ordering found in all the RCrSb$_3$ for R lighter than Gd. TbCrSb$_3$ shows a clear antiferromagnetic phase with $T_N=17$ K [Fig. 1(f)]. It is known that the ferromagnetic ordering in LaCrSb$_3$ is due to the ordering of the Cr atoms. According to Czachor, the antiferromagnetic order in RCrSb$_3$ compounds is due to the antiferromagnetic ordering of the $R^{3+}$ ions. In the present study, the evolution of an antiferromagnetic order from ferromagnetic order in RCrSb$_3$ as we changed R from La to Dy, is most likely due to favorable antiferromagnetic ordering of the $R^{3+}$ ions over the ferromagnetic order of the Cr atoms. This, most likely, has a connection with the changes in the lattice parameters due to the lanthanide contraction.

Magnetization data of DyCrSb$_3$ are plotted in Fig. 2(a). It shows that DyCrSb$_3$ orders antiferromagnetically below 12 K. Field-dependent magnetization data in the antiferromagnetic phase are plotted in Fig. 2(c) for DyCrSb$_3$. It clearly shows that the sample is antiferromagnetic below 12 K; more importantly, the sample transforms to a ferromagnetic phase with higher field showing a metamagnetic behavior of the sample. The critical field is found to be $H_C=1.9$ T at 5 K. The critical fields have been determined from several field-dependent magnetization measurements at different constant temperatures. An $H-T$ phase diagram is shown in the inset of Fig. 2(c). Hartjes et al.$^2$ also found a metamagnetic transition in PrV$_2$Sb$_3$ and NdV$_2$Sb$_3$. Since DyCrSb$_3$ showed metamagnetism, we also studied DyVSb$_3$. Figure 2(b) shows the magnetization of DyVSb$_3$ as a function of temperature. It is antiferromagnetic with $T_N=12$ K. Figure 2(d) shows the magnetization as a function of applied field at 5, 7, and 8 K where it is clear that DyVSb$_3$ also shows metamagnetic behavior. The inset of Fig. 2(d) shows the $H-T$ phase diagram for DyVSb$_3$.

Thermal expansion measurements were done on selected samples to determine the order of the ferromagnetic transitions and also to investigate any dramatic dimensional changes. Thermal expansion measurements were done on selected samples to determine the order of the ferromagnetic transitions and also to investigate any dramatic dimensional changes.
changes associated with the magnetic transitions. Thermal expansion data for LaCrSb$_3$, CeCrSb$_3$, and NdCrSb$_3$ are given in Figs. 3(a), 3(c), and 3(e), respectively. In the data for LaCrSb$_3$, a slope change occurs near the ferromagnetic transition which suggests a second-order transition. The CeCrSb$_3$ sample a slope change occurs near the ferromagnetic transition, which again suggests a second-order transition. A drop in sample dimension appears, near $T_N = 18$ K, the antiferromagnetic transition of NdCrSb$_3$, at higher temperatures, the data suggests a second-order ferromagnetic transition at $T_c = 141$ K.

Electrical resistance measurements on selected samples of RCrSb$_3$ were made to identify features in resistance which would correlate with the transitions observed in the magnetization measurements. Resistivity data on CeCrSb$_3$, PrCrSb$_3$, and NdCrSb$_3$ are shown in Figs. 3(b), 3(d), and 3(f), respectively. All samples show metallic character. In CeCrSb$_3$ the transition marked by an arrow corresponds to the ferromagnetic transition observed in the magnetization data. The two transitions found in the resistivity data of both PrCrSb$_3$ and NdCrSb$_3$ correspond to the antiferromagnetic and ferromagnetic transitions in the magnetization data.

**SUMMARY**

Magnetization, thermal expansion, and electrical resistivity measurements have been made on RCrSb$_3$ samples. RCrSb$_3$ samples show ferromagnetic order for R = La; antiferromagnetic and ferromagnetic orders for R = Ce, Pr, and Nd, and antiferromagnetic order for R = Gd, Tb, and Dy. The variety of magnetic order clearly depends on the size of the rare-earth elements. A drop in the sample dimension is found to occur near the Néel temperatures. The ferromagnetic transitions in RCrSb$_3$ are found to be second order. Transitions in electrical resistivity data corresponded to those in magnetization data.

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