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Magnetic properties of $RTSb_3$

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Magnetization, electrical resistivity, and thermal expansion measurements have been performed on polycrystalline $RTSb_3$ ($R=La, Ce, Pr, Nd, Gd, Tb, \text{ and } Dy$; $T=Cr, V$) samples in order to study their magnetic properties. Depending on the rare-earth element, $RTSb_3$ 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 R^{3+} and Cr^{3+} ions. As R is changed from La to Dy , the antiferromagnetic ordering of the R^{3+} ions becomes more prominent and predominate over the ferromagnetic ordering of Cr^{3+} for $R=Gd, Tb, \text{ and } Dy$. 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.

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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 $RTSb_3$ 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 chains¹ (seen in $CeCrSb_3$). Successive layering of such compound allows for the control of the magnetic aspect through interlayer coupling.

Previous results on the magnetic properties of the $RTSb_3$ 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.² The lowered Curie temperatures for $RTSb_3$ ($T=Cr$) 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 $LaTSb_3$ causes the structure of the compound to contract.¹ 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 $RTSb_3$ with $R=La, Ce, Pr, Nd, Gd, Tb, \text{ and } Dy$.

EXPERIMENTAL DETAILS

Polycrystalline $RTSb_3$ samples were synthesized by are melting of 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 superconduct-

ing 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 $RCrSb_3$ (with $R=La, Ce, Pr, Nd, Gd, \text{ and } Tb$) compounds as a function of temperature. Unless 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 $LaCrSb_3$ is ferromagnetic with a Curie temperature of 144 K. The transition temperature was determined from the first derivative of M vs T data. The T_c found by Hartjes *et al.*² for $LaCrSb_3$ is 125 K, which was determined from the lowest point of the $1/\chi$ vs T plot. In Fig. 1(b) $CeCrSb_3$ shows ferromagnetic ordering with $T_c=140$ K. This is much higher than that found by Hartjes *et al.*² The inset shows the magnetization data of $CeCrSb_3$ at 1 T. It shows that there is another ordering below 30 K. Interestingly, the $PrCrSb_3$ sample [Fig. 1(c)] shows antiferromagnetism below 16 K while it is ferromagnetic at higher temperatures with $T_c=142$ K. A comparison of the low-temperature ordering in $CeCrSb_3$ [the inset of Fig. 1(b)] with the low-temperature antiferromagnetic ordering in $PrCrSb_3$ suggests that in $CeCrSb_3$ there is an onset of an antiferromagnetic phase at low temperature. Temperature-dependent magnetization of the $NdCrSb_3$ [Fig. 1(d)] sample is similar to that of $PrCrSb_3$. For $NdCrSb_3$, 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.*²

We now turn our attention to new systems in the $RTSb_3$ series, namely, $GdCrSb_3$, $TbCrSb_3$, $DyCrSb_3$, and $DyVSb_3$.

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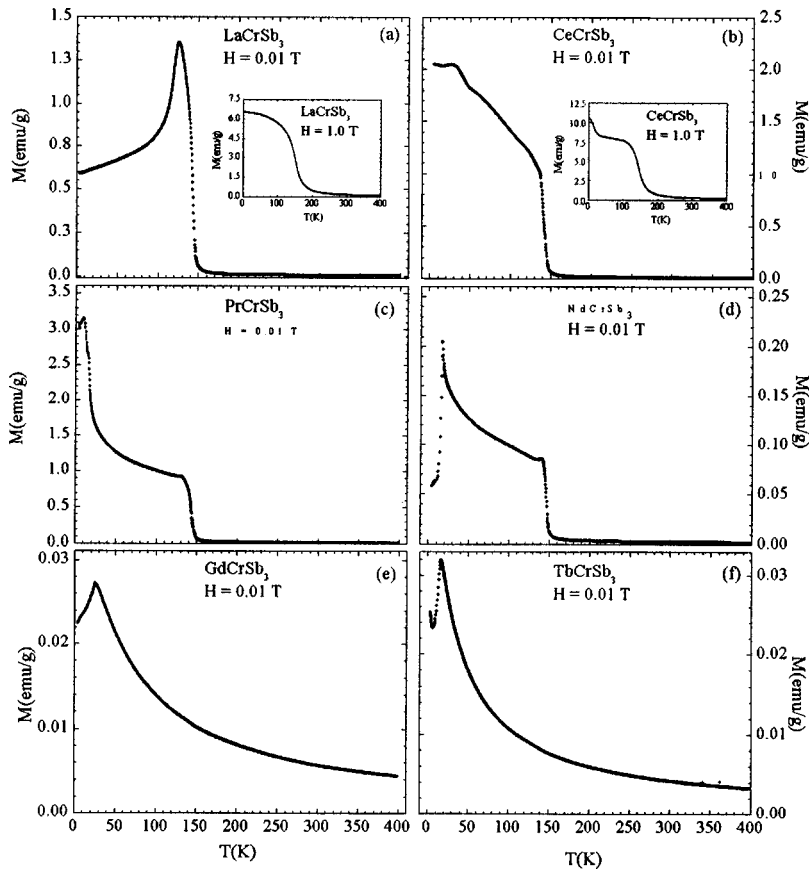


FIG. 1. Magnetization as a function of temperature of RCrSb₃ samples: (a) M vs T of LaCrSb₃ at 0.01 T; the inset shows the data taken at 1 T. (b) M vs T of CeCrSb₃ at 0.01 T; the inset shows the data taken at 1 T. (c) M vs T of PrCrSb₃ at 0.01 T; the inset shows the data taken at 1 T. (d) M vs T of NdCrSb₃ at 0.01 T. (e) M vs T of GdCrSb₃ at 0.01 T. (f) M vs T of TbCrSb₃ at 0.01 T.

In Fig. 1(e), GdCrSb₃ shows an antiferromagnetic ordering with $T_N=26$ K. GdCrSb₃ does not show the ferromagnetic ordering found in all the RCrSb₃ for R lighter than Gd. TbCrSb₃ shows a clear antiferromagnetic phase with $T_N=17$ K [Fig. 1(f)]. It is known that the ferromagnetic ordering in LaCrSb₃ is due to the ordering of the Cr atoms. According to Czachor,³ the antiferromagnetic order in RCrSb₃ compounds is due to the antiferromagnetic ordering of the R³⁺ ions. In the present study, the evolution of an antiferromagnetic order from ferromagnetic order in RCrSb₃ as we changed R from La to Dy, is most likely due to favorable antiferromagnetic ordering of the R³⁺ 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 DyCrSb₃ are plotted in Fig. 2(a). It shows that DyCrSb₃ orders antiferromagnetically below 12 K. Field-dependent magnetization data in the antiferromagnetic phase are plotted in Fig. 2(c) for DyCrSb₃. 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 PrVSb₃ and NdVSb₃. Since DyCrSb₃ showed metamagnetism, we also studied DyVSb₃. Figure 2(b) shows the magnetization of DyVSb₃ 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 DyVSb₃ also shows metamagnetic behavior. The inset of Fig. 2(d) shows the $H-T$ phase diagram for DyVSb₃.

Thermal expansion measurements were done on selected samples to determine the order of the ferromagnetic transitions and also to investigate any dramatic dimensional

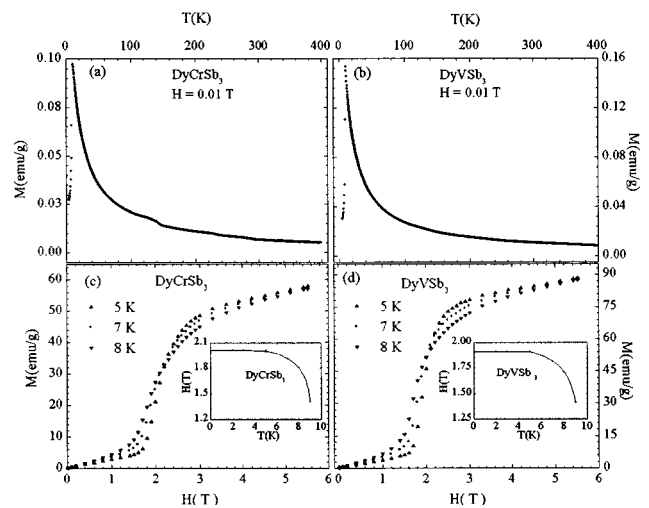


FIG. 2. Magnetization of DyCrSb₃ (a) and DyVSb₃ (b) samples as a function of temperature at an applied field of 0.01 T. Magnetization as a function of applied field at different constant temperatures of DyCrSb₃ (c) and DyVSb₃ (d) samples; the insets show the $H-T$ phase diagrams.

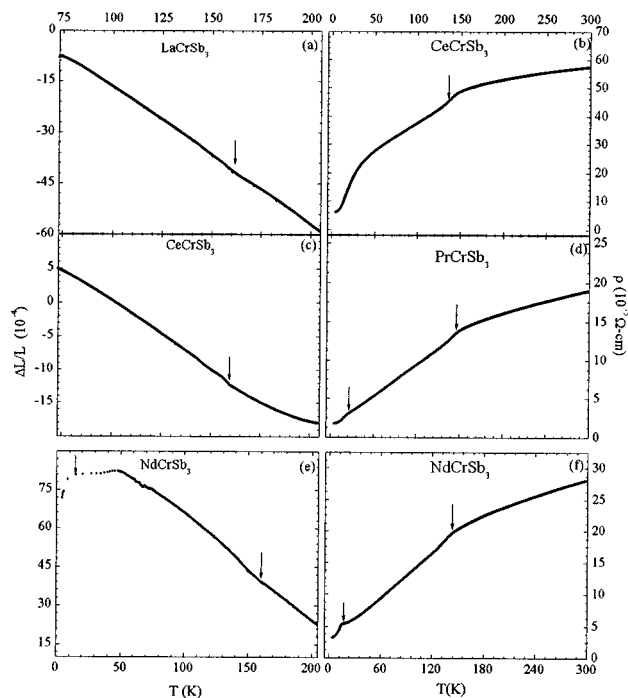


FIG. 3. Respective relative thermal expansivity $\Delta L/L$ of RCrSb_3 ($R=\text{La, Ce, Nd}$) are given in (a), (c), and (e) as a function of temperature. Electrical resistivity of RCrSb_3 ($R=\text{Ce, Pr, Nd}$) are shown in (b), (d), and (f), respectively, as a function of temperature.

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. In 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 RTSb_3 samples. RCrSb_3 samples show ferromagnetic order for $R=\text{La}$; antiferromagnetic and ferromagnetic orders for $R=\text{Ce, Pr, and Nd}$, and antiferromagnetic order for $R=\text{Gd, Tb, and Dy}$. 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.

ACKNOWLEDGMENT

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