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Magnetic and magneto-optical properties of $Mn_5(Ge_{1-x}M_x)_3$ alloys with M=Sn, Pb

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We report on the magnetic and magneto-optical properties of the intermetallic alloys $Mn_5(Ge_{1-x}M_x)_3$, where M=Sn, Pb. Generally, these alloys were ordered ferromagnetically at $T \geqslant 300$ K for $x<0.3$. The structure of $Mn_5(Ge_{1-x}Sn_x)_3$ remains in the hexagonal $Mn_5Si_3(hP16)$ phase up to $x=0.25$ and then changes to Ni_2In $(hP6)$ as x approaches 1. The magnetization and Curie temperature of these alloys is largest for small x and reach a minimum value near $x=0.5$. The room temperature Polar Kerr rotation, however, increases from $-0.06°$ at $x=0$ to maximum value of $-0.12°$ at $x=0.1$ before decreasing toward zero for $x>0.3$. The $Mn_5(Ge_{1-x}Pb_x)_3$ samples show a Mn_5Si_3 structure with ferromagnetic behavior for $x \leqslant 0.3$, and varying structure with rapidly decreasing magnetization above $x=0.3$. The magnetic behavior observed was complex for $x \geqslant 0.3$. The Kerr rotation of the Pb-doped alloys in the ferromagnetic region monotonically decreases from $-0.06°$ to zero at $x \approx 0.3$.

I. INTRODUCTION

Current research of materials for use as magneto-optic storage media focuses on ferromagnetic materials that have strong uniaxial anisotropy and Curie temperatures in the range of 30–200 °C.[1,2] The intermetallic compound Mn_5Ge_3 is ferromagnetic with a Curie temperature in the range of 27–47 °C. Favorable optical properties such as high optical absorption and large Kerr rotation might be expected largely due to the metallic character of the alloy and the high density of ferromagnetically aligned Mn^{2+} and Mn^{3+} ions present.[3] Panissod and his coworkers[4] have studied the $Mn_5(Ge_{1-x}Si_x)_3$ ferroantiferromagnetic transition as x changes from 0 to 1. They reported that Si can be substituted for Ge in Mn_5Ge_3 over the whole range of x and that these alloys are isostructural $(D8_8)$. The transition is from ferromagnetic for $x<0.85$ to antiferromagnetic for $x>0.85$. The aim of this work is to study the effects of heavy-element alloying on the magnetic properties and magnitude of the Kerr rotation (Θ_K). We report on the preparation and properties of the compounds $Mn_5(Ge_{1-x}M_x)_3$ with M=Sn, Pb.

II. EXPERIMENTS

Samples were prepared from 99.995 at. % pure Mn, 99.99 at. % pure Ge, and 99.9999 at. % pure Sn or 99.999 at. % pure Pb. Stoichiometric amounts were alloyed by arc melting in an argon atmosphere. In order to obtain homogeneous alloys, each sample was melted four to five times. Mass losses during melting totaled about 2% for $Mn_5(Ge_{1-x}Sn_x)_3$ and about 3% for $Mn_5(Ge_{1-x}Pb_x)_3$. The samples were then annealed in vacuum at 700 °C for 10 days.

The structure of the samples was analyzed by x-ray diffractometry. Their magnetization and coercivity were measured using an alternating gradient force magnetometer (AGFM) and superconducting quantum interference device (SQUID). The Curie temperatures were measured by both a high-temperature Faraday balance magnetometer and AGFM at $H=500$ Oe. The Polar Kerr rotation was determined by a home-made apparatus[5] at room temperature and fields up to 8 kOe.

III. RESULTS AND DISCUSSION

A. Crystallographic data

Figure 1 shows x-ray diffraction measurements on representatives of the $Mn_5(Ge_{1-x}Sn_x)_3$ samples. $Mn_5(Ge_{0.90}Sn_{0.10})_3$ [Fig. 1(a)] is found to have a $Mn_5Si_3(hP16)$ type structure, as does $Mn_5(Ge_{0.70}Sn_{0.30})_3$ [Fig. 1(b)] but with a $Ni_2In(hP6)$ structure phase also beginning to appear. The $Mn_5(Ge_{0.30}Sn_{0.70})_3$ sample [Fig. 1(c)] shows the $Ni_2In(hP6)$ structure. Only small amounts of Mn oxides show up in the x-ray spectra. It has also been found by others that Mn_5Ge_3 has a Mn_5Si_3 structure[3] and Mn_5Sn_3 has a Ni_2In structure.[6] When Sn is substituted for Ge in Mn_5Ge_3 [i.e., $Mn_5(Ge_{1-x}Sn_x)_3$] it can be expected that for small x the compounds will maintain the Mn_5Si_3 structure and that for large x the compounds should form the Ni_2In structure. In the middle range these structures could be mixed and show various lattice distortions. Since the Sn atomic diameter is larger than that of Ge, when Sn is substituted for Ge in Mn_5Ge_3 the lattice parameter of the crystal changes. Table I gives the variations in the lattice parameters measured in our samples.

Figure 2 shows x-ray diffraction measurements on some of the $Mn_5(Ge_{1-x}Pb_x)_3$ samples. We find that, as expected, $Mn_5(Ge_{0.95}Pb_{0.05})_3$ [Fig. 2(a)] has the $Mn_5Si_3(hP16)$ structure. This structure type becomes unstable at $x>0.3$. $Mn_5(Ge_{0.50}Pb_{0.50})_3$ [Fig. 2(b)] shows the Ni_3Sn $(hp8)$ type structure, which is predominant for this composition. $Mn_5(Ge_{0.30}Pb_{0.70})_3$ [Fig. 2(c)] mainly shows the Ni_3Sn $(hp8)$ type structure but broad peak is found at a position expected from a pure tetragonal Mn phase. We also note that for x larger than 0.5 the desired compounds were difficult to form. This was especially noted for Mn_5Pb_3, which often resulted

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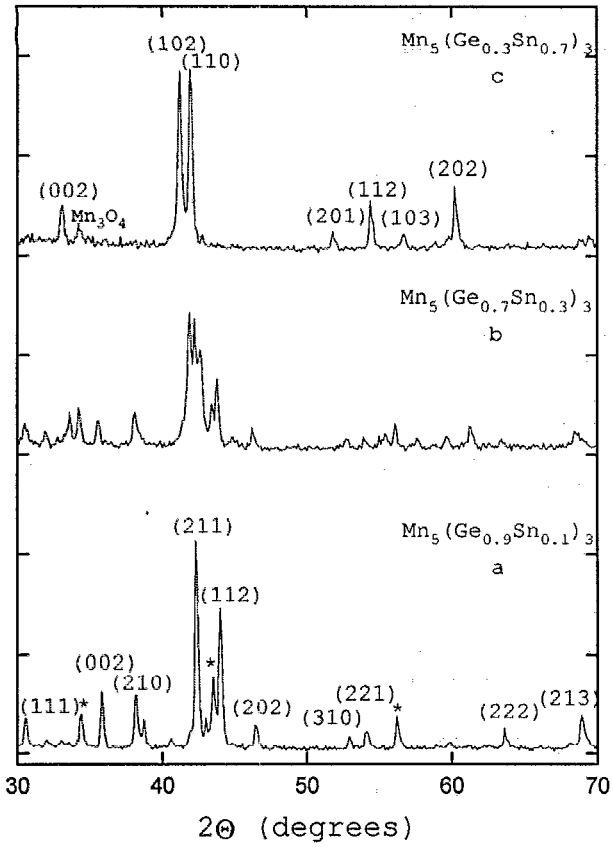

FIG. 1. X-ray diffraction patterns for several compositions of $Mn_5(Ge_{1-x}Sn_x)_3$: (a) $x=0.1$ with the Mn_5Ge_3 ($hP16$ Mn_5Si_3) spectrum labeled. The asterisks denote weak peaks attributed to Mn_2Ge ($hP6$ Ni_2In). (b) $x=0.3$. The spectrum shows peaks for both Mn_5Ge_3 ($hP16$ Mn_5Si_3) and Mn_2Ge ($hP6$ Ni_2In), with the former being dominant. (c) $x=0.7$ with peaks for $Mn_{1.77}Sn$ ($hP5.54$ Ni_2In) labeled.

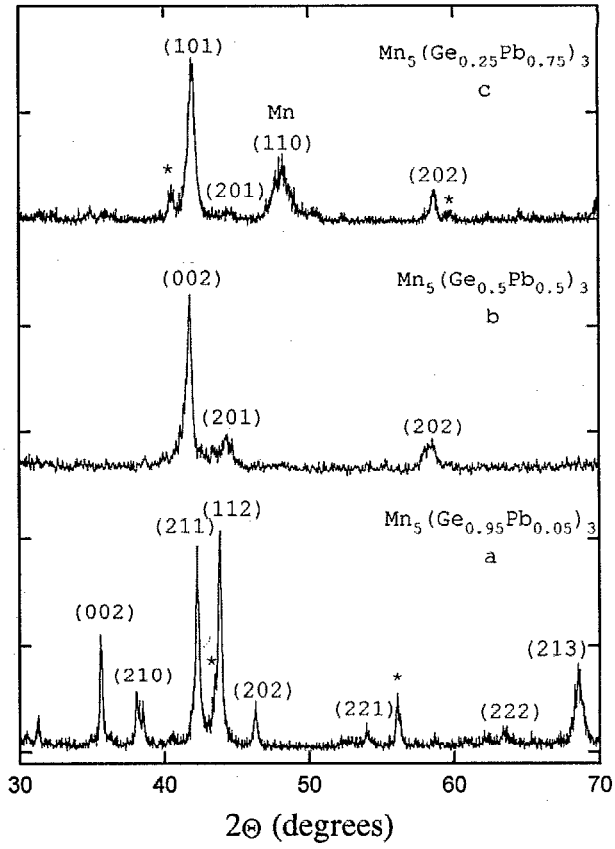

FIG. 2. X-ray diffraction patterns for several compositions of $Mn_5(Ge_{1-x}Pb_x)_3$: (a) $x=0.95$ with labels corresponding to the Mn_5Ge_3 ($hP16$ Mn_5Si_3) spectrum. The asterisks denote weak peaks attributed to Mn_2Ge ($hP6$ Ni_2In). (b) $x=0.5$. The labels correspond to the Mn_3Ge ($hP8$ Ni_3Sn) spectrum. (c) $x=0.7$ with the labeled peaks belonging to the Mn_2Ge ($hP6$ Ni_2In) structure.

in a fcc Pb structure with the main peak from the β-Mn spectrum also present. Mn has a smaller atomic diameter than Pb so it is expected that Mn_5Pb_3 in a fcc phase have a smaller lattice parameter than the pure fcc Pb lattice. These effects of alloy substitutions are confirmed by our experimental results. As shown in Table I, we found that the cubic lattice parameter a changes from 4.962 Å for the pure Pb crystal to 4.955 Å for the Mn_5Pb_3 alloy.

TABLE I. Structural lattice parameters for the relevant compositions of $Mn_5(Ge_{1-x}M_x)_3$ for M=Sn, Pb, and the average magnetic moment per Mn atom at $T=5$ K. Data are only shown for samples from which it could be clearly calculated.

Composition	Lattice parameter a	Lattice parameter c	Average moment per Mn atom (μ_B)
Mn_5Ge3	7.193	5.069	2.52
$Mn_5(Ge_{0.95}Sn_{0.05})_3$	7.196	4.990	2.52
$Mn_5(Ge_{0.90}Sn_{0.10})_3$	7.176	5.061	2.23
$Mn_5(Ge_{0.85}Sn_{0.15})_3$	7.116	4.978	2.23
$Mn_5(Ge_{0.80}Sn_{0.20})_3$	7.172	5.036	2.13
Mn_5Ge_3	7.193	5.069	2.52
$Mn_5(Ge_{0.95}Pb_{0.05})_3$	7.179	5.048	1.42
$Mn_5(Ge_{0.90}Pb_{0.10})_3$	7.164	5.055	2.16
$Mn_5(Ge_{0.85}Pb_{0.15})_3$	7.168	5.039	2.10
$Mn_5(Ge_{0.80}Pb_{0.20})_3$	6.755	5.212	1.23
Mn_5Pb_3	4.955	4.955	0.01
Pb	4.963	4.963	0.00

B. Magnetic properties

Figure 3(a) shows the compositional dependence of the magnetization measured at $T=5$ K and a maximum field of 50 kOe with a SQUID magnetometer. The Curie temperatures for the compounds studied are shown in Fig. 3(b). All of the $Mn_5(Ge_{1-x}Sn_x)_3$ alloys made have a ferromagnetic transition and for $x\leq 0.3$ they are ferromagnetic at room temperature. The magnetization at $T=5$ K first generally decreases with increasing x, and then shows a slow increase above $x=0.5$. The coercivity however is maximized for $x=0.5$, reaching 1.2 kOe at $T=5$ K. The Curie temperature initially remains nearly constant, then decreases with increasing x beyond $x=0.3$, reaching a minimum at $x=0.5$, after which it increases again as x goes to 1.0.

It can be seen in Table I that as x increases, both the lattice parameter a and the average moment per Mn atom generally decrease. This is understandable since the hybridization of Mn $3d$ electronic states normally increases as the interatomic separation becomes smaller as given by the quantum theory.

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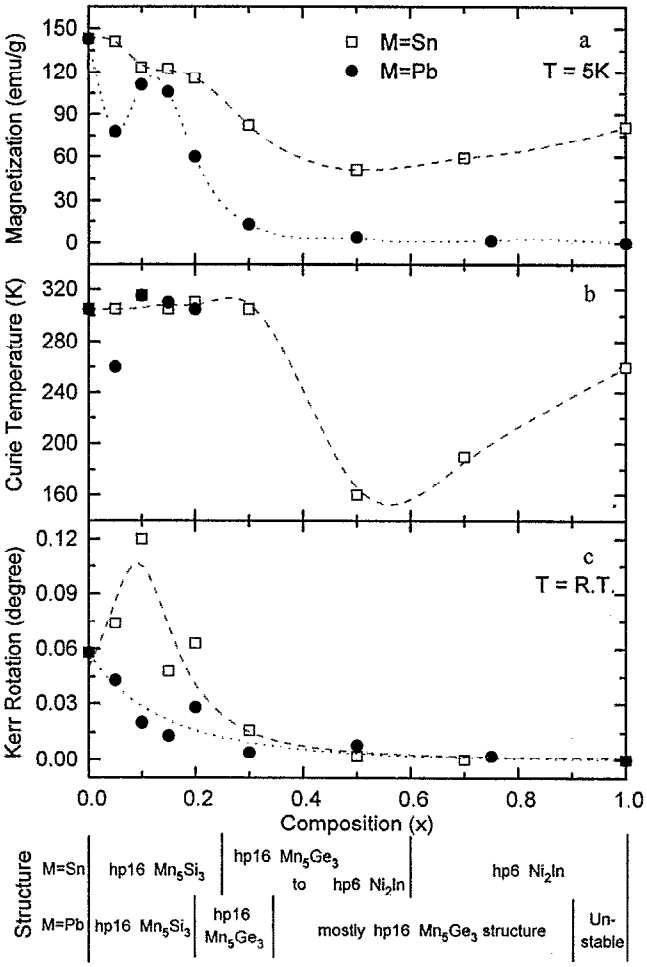

FIG. 3. Magnetic properties verses composition for $Mn_5(Ge_{1-x}M_x)_3$, M=Sn, Pb: (a) SQUID measurements of magnetization at $T=5$ K and $H=50$ kOe. (b) Curie temperature measurements from Faraday balance magnetometer and M vs H loops with fields up to 500 Oe. (c) Kerr rotation measurements at room temperature and fields up to 8 kOe. The curves are only meant to serve as a reference to the general trends noted. The various structure regions for each sample set are also shown for reference.

The $Mn_5(Ge_{1-x}Pb_x)_3$ alloys are ferromagnetic at room temperature only for $x \leqslant 0.2$. The magnetization and Curie temperature show a minimum at $x=0.05$, for reasons that are not presently clear. When $x>0.1$ both of these properties decrease as x increases. The explanation is the same as given for the $Mn_5(Ge_{1-x}Sn_x)_3$ alloys concerning the hybridization of the Mn $3d$ states. When $x>0.3$ the Pb substituted alloys show a complex magnetic behavior. For example, the $Mn_5(Ge_{0.25}Pb_{0.75})_3$ sample measured at $T=5$ K and up to $H=50$ kOe has a small (about 1.5 emu/g) magnetization which is unsaturated, and a coercivity of about 11 kOe, while at room temperature it still has a coercivity of about 2 kOe.

C. Magneto-optic properties

Figure 3(c) shows the relationship of the magnitude of the Polar Kerr rotation to the composition, measured at room temperature in fields up to 8 kOe. The sign of the rotation is negative for all of the samples studied. For $Mn_5(Ge_{1-x}Pb_x)_3$, the Kerr rotation monotonically decreases from the value for the parent alloy Mn_5Ge_3 with increasing x. In agreement with the magnetization at room temperature with an applied field of 8 kOe the Kerr rotation of the Sn substituted compounds increased slightly for $x<0.15$. The peak value of $\theta_K=0.12°$ at $x=0.1$ is enhanced above what would be expected from a direct relationship between θ_K and M. Beyond this peak the Kerr rotation drops off, going to zero rapidly outside the single phase region at $x=0.25$. The Kerr rotation values below $x=0.4$ for both cases was measured very near the Curie temperature in both series of samples which would adversely affect the magnitude of the rotation due to decreased magnetic magnetizations. However, the Polar Kerr rotation is an intrinsic quantity that depends on factors such as composition, crystal structure, and electronic structure. It is interesting that there is an enhanced maximum in Θ_K for the Sn case that apparently occurs within a single phase region. Presumably this reflects subtle changes in the electronic structure. This indicates the need for at least a spin-polarized calculation on the parent Mn_5Ge_3 compound.

IV. CONCLUSION

The magnetic and magneto-optical properties strongly depend on the alloy structure. Our $Mn_5(Ge_{1-x}Pb_x)_3$ samples showed the Mn_5Si_3 structure and were ferromagnetic for $x \leqslant 0.3$, exhibiting complex magnetic behavior for $x>0.3$. All of the $Mn_5(Ge_{1-x}Sn_x)_3$ alloys were also ferromagnetic, but the low-temperature magnetization showed a minimum value at $x=0.5$. Only the substitution of Sn for Ge in Mn_5Ge_3 allowed the alloy to both keep the Mn_5Si_3 structure and increase the Kerr rotation.

In general the presence of heavy elements is necessary for large spin-orbit interactions, which can aid in producing a large Kerr effect; in the case of these compounds there is a compensating effect on alloying with Sn or Pb that reduces the strength of the magnetic coupling, as evidenced by a reduction in M_s and T_c. Thus, in these compounds, any tendency for Kerr-effect enhancement is overwhelmed by the destruction of the ordered magnetic state.

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