Magnetoresistance of $SmMn_2Ge_2$: A layered antiferromagnet

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(Received 21 October 1992)

The behavior of the magnetoresistance in $SmMn_2Ge_2$—a layered intermetallic compound—is analogous to the giant magnetoresistance recently reported for artificial metallic multilayers. A moderate magnetic field (2–15 kOe) is sufficient to switch $SmMn_2Ge_2$ from the antiferromagnetic (metamagnetic) state to saturation, at a temperature of ~100 K, and this results in a resistance change of 4–8 %. The study of spin-dependent scattering in this and similar layered structures, in which nature has conspired to make essentially perfect interfaces, serves as a complement to the study of the artificial structures.

Recent interest[1–13] in the magnetoresistance of artificial magnetic multilayers has been stimulated by the observation that antiferromagnetically (AFM) coupled layers can show a dramatic decrease in resistivity when the layers are forced into ferromagnetic (FM) alignment by the application of an external magnetic field. The observed magnetoresistance, defined as $(R_{max}-R_{min})/R_{min}$, can be as high[6] as 80% at 4.2 K, with a saturation field of roughly 7 kOe. Theoretical interpretations[11,12] have focused on spin-dependent scattering both at layer boundaries and within each layer, resulting in the prediction that the enhancement of the magnetoresistance depends in part on the presence of surface roughness. Fullerton *et al.*[13] have reported observing this effect in sputter-deposited films.

While artificial multilayers are convenient for many investigations, naturally layered compounds are an important alternative for delineating fundamental issues concerning transport in two-dimensional (2D) magnetic materials. We have investigated the behavior of $SmMn_2Ge_2$, which forms in the $ThCr_2Si_2$ crystal structure in which the layering along the c axis follows the sequence . . . Sm-Ge-Mn_2-Ge. . . . The Mn layers are square planes with Mn-Mn separations of 2.87 Å; adjacent planes are in registry along the stacking direction, with an interplane spacing of 5.45 Å. Thus the Mn-Mn interactions are highly anisotropic. This material is a reentrant ferromagnet,[14–17] i.e., at $T=0$ it is ferromagnetic, at ~97 K it undergoes a first-order phase transition into an antiferromagnetic configuration, and at ~153 K undergoes another first-order transition into a ferromagnetic state with a Curie temperature[15] of 348 K. The two first-order transition temperatures are sensitive to pressure,[17] and can also be altered by partial substitutions of Gd (Ref. 18) or Y (Ref. 19).

Neutron-diffraction data for $TbMn_2Ge_2$ and YMn_2Ge_2, as well as a number of RMn_2Si_2 compounds show[20] that the magnetic moments within the Mn planes are parallel and directed along the c axis, while alternate Mn planes have antiparallel moments. It is inferred that this case obtains for $SmMn_2Ge_2$ as well.[14] (Note that in the artificial multilayers the easy axis is in the plane of the films.) The field necessary to switch the moments into ferromagnetic alignment can be fairly small (2–15 kOe); this is commonly referred to as metamagnetism. We have found that the difference in resistance between the ferro- and antiferromagnetic configurations is about 4–8 % in this material, and since it can be switched by application of a magnetic field, represents a magnetoresistance comparable to that observed in artificial multilayers.

In the present work we investigated the properties of $SmMn_2Ge_2$ crystals grown in an indium flux,[21] and crystals grown by arc melting; the arc-melt crystals were superior so we report exclusively on these. Buttons with the composition $SmMn_{2.02}Ge_2$ (to compensate for a small Mn loss) were arc-melted under Ar. They were then annealed for 10 days at 800 °C to improve homogeneity and crystallinity. The buttons were single-phase by powder x-ray diffraction with refined tetragonal cell parameters $a_0=4.065(1)$ and $c_0=10.899(4)$ Å. They were crushed to free small (typically $0.8\times0.4\times0.09$ mm^3) crystals, which were mounted for transport measurements with contacts in the van der Pauw or in-line configuration. The results in both cases were the same. The resistance was measured using a dc current of 1 mA with the magnetic field applied along the c axis. The absolute c-axis resistivity was inferred at room temperature using the Montgomery method.[22] The shape of the sample was slightly irregular, leading to some inaccuracy in the absolute scale of the resistivity; relative changes are of course much easier to measure precisely. The ac susceptibility was measured in a 111.1 Hz, 10 Oe rms field using a Lakeshore 7000 susceptometer. The dc magnetization was measured using a vibrating sample magnetometer, and is presented with the background slope (sample holder diamagnetism) subtracted digitally.

Figure 1(a) shows the temperature-dependent ac susceptibility of a typical crystal. The magnetic and electrical properties of selected crystals were found to be reproducible. The first-order transitions into and out of the AFM state are clearly seen in Fig. 1 at roughly 95 and 150 K, respectively. A small hysteresis, ~4 K, can be seen as well. The in-plane resistivity, ρ_a, is shown in Fig. 1(b), where the magnetic transitions are clearly delineated. At 300 K we find $\rho_a=260$ $\mu\Omega$ cm. The overall temperature dependence is typical of ferromagnetic materials, being accurately quadratic; this suggests spin-wave

scattering.[23] Figure 1(c) presents the same resistivity data with the quadratic temperature dependence in the ferromagnetic state subtracted. In this plot the zero-field resistive transitions are clear and the magnitude of the normalized resistance jump $\Delta\rho_a/\rho_a$ at 97 and 153 K are seen to be nearly identical. The c-axis resistivity of this sample at 300 K is 6700 $\mu\Omega$ cm, implying an anisotropy of about 25 with respect to the a axis, validating our description of this material as a layered magnetic material.

While the AFM state has the lowest energy at zero field for $97 < T < 153$ K, the ferromagnetic state will have a lower energy in a sufficiently high field. Thus when the sample is held at a temperature between 93 and 156 K, it can be magnetically switched into the FM state (reversibly if the temperature is between 101 and 150 K). This is demonstrated in Fig. 2, for which the sample was held at roughly 104 K (indicated by the arrows in Fig. 1). Figure 2(a) shows the magnetization curve for the sample, showing a switching field $H_{s1}=8$ kOe, and a return field $H_{s2}=3$ kOe. Similar curves have been found for certain molecular-beam-epitaxy (MBE) grown multilayer films[1,24] but typical multilayer films do not show such clear metamagnetic behavior. The squareness of the displaced loops suggests that the easy axis in the AFM and FM states is along the direction of the magnetic field, i.e., the c axis. Note, however, that in the ferromagnetic state below 95 K the easy axis is not along the (001) direction but along the (110) directions. The value of the switching field is temperature dependent and somewhat sample dependent, as is the hysteresis $H_{s1}-H_{s2}$.

Figure 2(b) shows the in-plane transverse magnetoresistance, normalized to the zero-field value. The resistance is seen to exhibit a 7.5% increase on switching from the AFM to the FM state. This value is higher than the 5% jumps seen in the $\rho(T,H=0)$ curve of Fig. 1(b). The switching fields are somewhat greater than those seen in Fig. 2(a), because the temperature was slightly higher for the resistance measurements.

A lattice contraction of 0.2% in the a-axis and an expansion of $\sim 0.1\%$ in the c axis associated with the AFM phase of $SmMn_2Ge_2$ was inferred from x-ray-diffraction measurements as a function of temperature.[16] We might expect that the field-driven phase transition would be accompanied by a similar structural change. Due to the experimental difficulty involved in x-ray measurements at low temperatures and in a magnetic field, we used an alternative indicator, namely, the macroscopic strain of a polycrystalline sample. Figure 3(a) shows the temperature dependence of the strain observed in a polycrystalline sample ($5\times5\times1$ mm^3), obtained by differential strain-gauge measurement with respect to the strain of a (1102)-face sapphire reference. The jumps at the AFM-FM transitions and the overall thermal expansion are

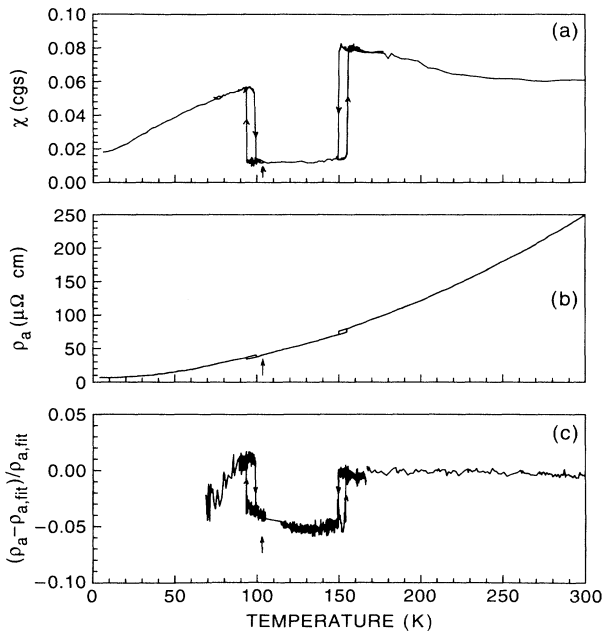

FIG. 1. (a) ac susceptibility of an $SmMn_2Ge_2$ crystal, measured with a 10 Oe, 111.1 Hz field applied along the c axis of the crystal. Below 97 K the material is ferromagnetic, between 97 and 153 K it is antiferromagnetic, and between 153 and 348 K it is ferromagnetic; the last is the Curie temperature. (b) Zero-field resistivity of the same crystal, measured perpendicular to the c axis (ρ_a) as a function of temperature. Transitions in the resistivity at 97 and 153-K are clearly evident. (c) Normalized residuals obtained by subtracting a quadratic fit from the resistivity data above. This accentuates the zero-field resistive transitions associated with the antiferromagnetic phase. The two normalized jumps have a nearly identical magnitude.

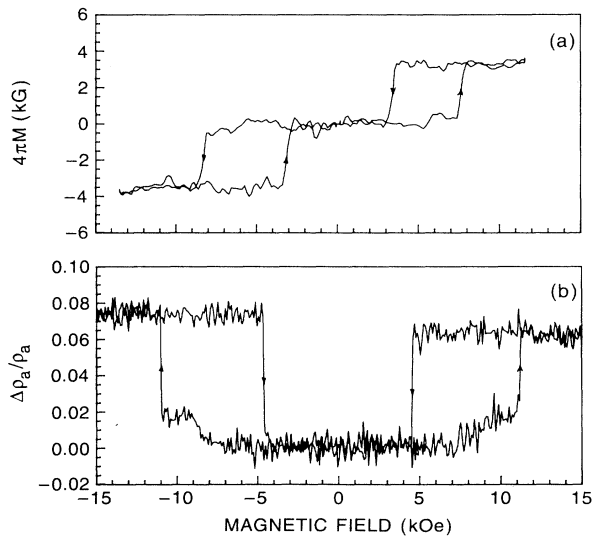

FIG. 2. (a) M-H curve of $SmMn_2Ge_2$ at ~ 104 K, as measured with a vibrating-sample magnetometer (VSM). A background linear slope due to the diamagnetic sample holder has been subtracted. The magnetic field is applied along the c axis. (b) Magnetoresistance of the same crystal as in (a), measured in the basal plane with the field parallel to the c axis at a temperature of 104.4 K. The large jumps correspond to the transition from the antiferromagnetic to ferromagnetic state. The curvature in the low-resistance curve is tentatively attributed to inhomogeneous stress introduced by the Ag-epoxy contacts.

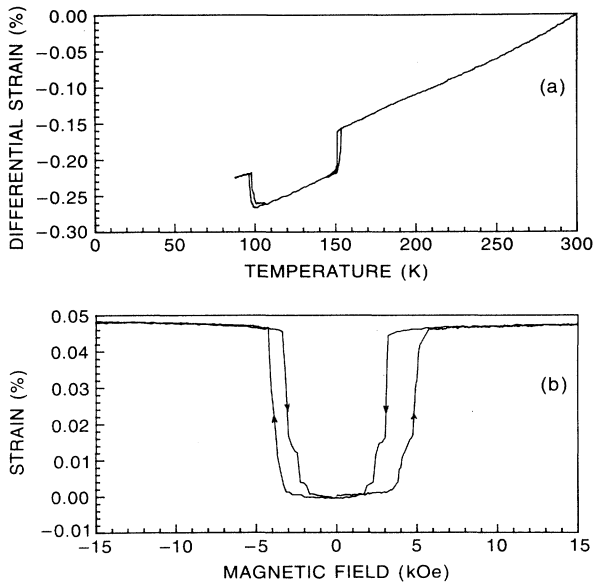

FIG. 3. (a) Temperature dependence of the strain of a polycrystalline sample of $SmMn_2Ge_2$, measured with respect to a (1102)-face sapphire reference. (b) Strain in the same sample measured at 104.4 K as a function of field. The magnitude of the jump at the field-induced AFM-FM transition is the same as in (a).

easily resolved, although the absolute magnitude is decreased due to the polycrystalline nature of the sample. The strain-gauge technique also allows straightforward measurement of the isothermal changes in the lattice constant as a function of magnetic field. Figure 3(b) shows that the strain associated with the field-induced AFM-FM transition at 104.4 K is identical to that measured in zero field as a function of temperature.

We also investigated the behavior of our sample at 145 K, and found it to be qualitatively similar to the behavior shown in Fig. 2. That is, close to the AFM-FM transition the spin-flip field is well below 10 kOe, and the magnetoresistance is moderately large ($\sim 8\%$) and consistent with the data of Fig. 1.

The magnetoresistance of $SmMn_2Ge_2$ may arise from a number of sources. First, there may be a change in the Fermi surface associated with the AFM-FM transition. While the changes in lattice constants at the first-order transitions cannot account directly for the magnitude of the observed resistivity change, they may have subtle effects on the band structure. Calculations of the band structure of $Mn_{2-x}Cr_xSb$, for example, have been used to interpret the 100% resistance change at the AFM-ferrimagnetic transition in that material,[25] which has, however, a less anisotropic layered structure than $SmMn_2Ge_2$. There the increased resistance in the AFM state was attributed to an increase in the effective mass due to additional electron orbits at the Fermi surface which have a particularly low dispersion. Other sources for the magnetoresistance have been suggested for the case of, for example, UNiGa, a moderately anisotropic compound in which a magnetoresistance of 650% at 4.2 K was measured.[26] In that material the magnetoresistance was attributed to the possibility of the disappearance of magnetic superzones in the ferromagnetic state, or else decoupling of conduction electrons from f electrons.

These band-structure effects are interesting if not novel, and may play a role in the behavior of $SmMn_2Ge_2$. But in this highly anisotropic layered structure we have the additional possibility of spin-dependent interlayer magnetic scattering as proposed originally by Binasch *et al.*[1] This mechanism is only effective in the AFM state and should lead to a higher resistance compared to the FM state, as is observed both in artificial multilayers even where the thickness of the magnetic layers and nonmagnetic spacers (a few atoms each) approaches the values for $SmMn_2Ge_2$. Yet, as we have shown, the a-axis resistivity of $SmMn_2Ge_2$ is lower in the AFM state. Evidently the interlayer magnetic scattering channel is overwhelmed by some other effect. This only demonstrates that interlayer scattering is small, not that it is absent. It is not an entirely surprising result, given that this channel is expected to depend on surface roughness, which is notably absent in our samples. Further studies with deliberately included disorder should elucidate this point, and may be complemented by studies of related 2D magnetic compounds.

$SmMn_2Ge_2$ shows promise as a model system for understanding artificial multilayers, because it exhibits a very clean metamagnetic transition, and because intermixing between layers (unavoidable in artificial multilayers[27]) can be controlled. Given the wide range of layered structures possible, we suggest that much of the rich variety of behavior and novel physics in layered magnetic materials can be conveniently studied using appropriate bulk materials.

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