Negative giant longitudinal magnetoresistance in NiMnSb/InSb: Interface effect

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We report on the electrical and magnetotransport properties of the contact formed between *n*-type degenerate InSb (100) substrates and polycrystalline NiMnSb thin films grown using pulsed laser deposition. Negative giant magnetoresistance is observed when the external magnetic field is oriented parallel to the in-plane current direction. We attribute the observed phenomenon to magnetic precipitates that are formed during the magnetic film deposition and are confined to a thin layer at the interface. The evidence for the formation of a thin interfacial layer is obtained through x-ray reflectivity measurements.

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I. INTRODUCTION

There has been an extensive interest in the giant magnetoresistance (GMR) effect since its discovery in Fe/Cr multilayers[1] and subsequently in many other multilayers[2,3] such as Fe/Cu and Co/Cu and sandwich structures such as Co/Au/Co.[4] Much of this research interest has been driven by the important applications of the GMR effect in the read heads of modern hard drives and in nonvolatile magnetic random access memories. The GMR effect has also been observed in heterogeneous alloys with ferromagnetic granules embedded in a nonmagnetic metallic matrix;[5,6] however, these materials have not been able to produce the high GMR ratios found in the multilayer counterparts. This investigation has been extended to granular systems consisting of ferromagnetic clusters embedded in semiconductors. For example, negative magnetoresistance has been reported in GaAs films containing MnAs ferromagnetic clusters[7] and, more recently, in thin film magnetic systems consisting of nanoscale MnGe ferromagnetic clusters embedded in a Mn_xGe_{1-x} dilute ferromagnetic semiconductor.[8] The observation of such effects in semiconductor systems containing ferromagnetic clusters is particularly important in the microelectronics industry, since semiconductors are integratable and allow for the possibility to fabricate chips which can utilize both the charge and the spin of electrons.

Recently, we reported on the successful growth of stoichiometric polycrystalline NiMnSb thin films on InSb substrates using pulsed laser deposition (PLD).[9] Our films exhibit a saturation magnetization of $4\mu_B$/formula unit at 5 K and a magnetization temperature dependence that follows Bloch's $T^{3/2}$ law, consistently with the expected half-metallic behavior. In this paper we report on an unusual negative GMR effect observed in the NiMnSb/InSb system when the magnetic field is applied parallel to the in-plane current direction. A similar effect is also observed when a ferromagnetic (FM) film of Ni is deposited on InSb. On the other hand, no negative GMR effect is displayed when the deposited film is nonmagnetic. Additionally, given that none of the two constituents of the FM/InSb system demonstrates any negative magnetoresistance effect, we argue that the GMR effect is due to magnetic precipitates which are formed at the interface during the growth of the magnetic films. These magnetic precipitates align their magnetic moments in the direction of the external magnetic field and thus the spin-dependent scattering of the electrons is reduced. Evidence for the formation of the interfacial layer containing the magnetic precipitates is obtained by x-ray reflectivity (XRR) measurements. The strong anisotropy of the investigated system's magnetoresistance makes it a good candidate for use in magnetic field sensors.

II. EXPERIMENTAL DETAILS

NiMnSb films were grown by the conventional PLD method onto heated ($T_s \sim 200\,°C$) *n*-type InSb (100) substrates. Further details of the growth conditions of the films and their structural and magnetic properties can be found elsewhere.[9] In order to investigate the electrical contact formed between the NiMnSb film and the InSb substrate, we performed detailed resistance and magnetoresistance (MR) measurements on both the NiMnSb/InSb system and the bulk InSb substrates. It is important to note that in the former case, the electrodes for the four-terminal measurements were placed on the top NiMnSb layer (current-in-plane configuration). The electrical resistance was measured by the standard four-probe ac method in the temperature range $5 < T < 300$ K and in the magnetic field interval 0–7 T. MR measurements were taken for both positive and negative magnetic fields in order to eliminate effects due to probe misalignment. XRR measurements were carried out using a

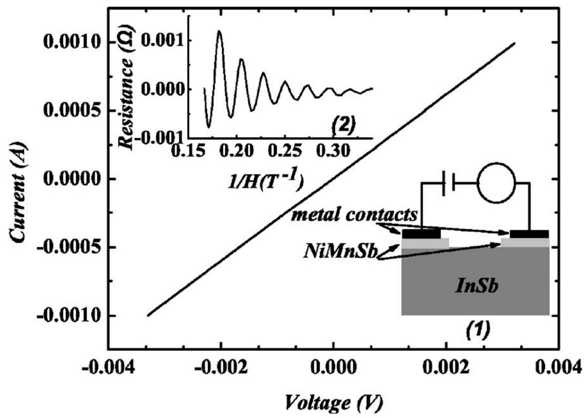

FIG. 1. *I-V* characteristic for the NiMnSb/InSb system at 5 K. Inset 1: Schematic of the setup used for the *I-V* measurements. Inset 2: The oscillating part of the transverse MR as a function of $1/H$ for the InSb substrate at 5 K after subtracting a background. As background we used the MR at 50 K (shown in Fig. 3 below), which was the same curve in amplitude as the MR at 5 K, without the oscillations. We fitted the background with a polynomial of the form Ax^2+Bx+C.

reflectometer developed in house; further details have been published elsewhere.[10]

III. RESULTS AND DISCUSSION

As shown in Fig. 1 the *I-V* characteristic curve obtained at 5 K indicates that the contact between NiMnSb and InSb is Ohmic. We note that the temperature dependence of the electrical resistance (not shown) of the *n*-type degenerate InSb substrates exhibits metalliclike behavior.

MR measurements of the *n*-type InSb (100) substrate revealed the presence of Shubnikov–de Haas (SdH) oscillations for magnetic fields applied both perpendicular and parallel to the crystal surface. These magneto-oscillations are due to the Landau quantization of the three-dimensional electronic density of states.[11] The MR oscillations are periodic in $1/H$ (inset 2, Fig. 1). From the period $\Delta(1/H)=0.023T^{-1}$ of the oscillations in the transverse MR, we calculated the electron density for the InSb substrate, $n=1.6\times10^{18}$ cm^{-3}, via the formula[12] $\Delta(1/H)=(2e/\hbar)(3\pi^2 n)^{2/3}$. Such a high value of n explains the metalliclike behavior of InSb and the Ohmic behavior of the contact formed between NiMnSb and InSb. Surprisingly, SdH oscillations were also observed in the MR of the NiMnSb/InSb system, i.e., following the deposition of a 600-nm-thick NiMnSb layer on top of the InSb substrate. This is further evidence of the good Ohmic behavior of the NiMnSb-InSb electrical contact.

Apart from the oscillating part of the MR, the background of the MR gives some important information about the electron transport in these systems. In both InSb and NiMnSb/InSb systems the transverse MR is found to be positive and quadratic in H [Fig. 2(c)]. This is the characteristic of the geometric magnetoresistance which is due to the enhanced scattering of the electrons that are forced in circular orbits by the applied magnetic field.

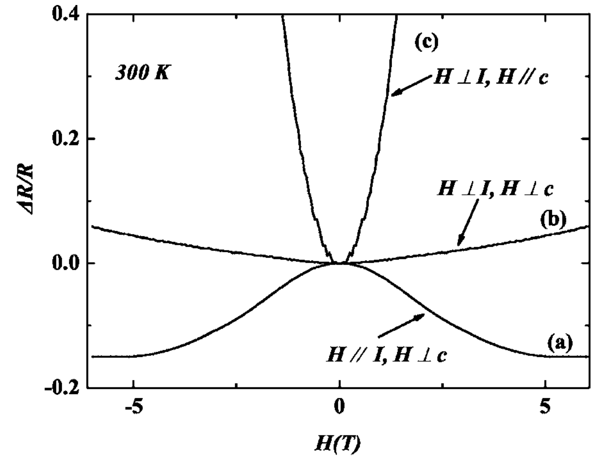

FIG. 2. Plots of the ratio $\Delta R/R$ as a function of the magnetic field (H) obtained at 300 K for magnetic field applied (a) in plane ($H\perp c$) and parallel to current flow ($H\parallel I$), (b) in plane ($H\perp c$) and perpendicular to current flow ($H\perp I$), and (c) perpendicular to plane ($H\parallel c$) and to current flow ($H\perp I$).

The longitudinal MR of the NiMnSb/InSb system, with H applied in plane and parallel to the current, corresponding to zero Lorentz force on the charge carriers, exhibits the most interesting behavior. Apart from the low-temperature oscillations, there is a negative MR background which persists almost unchanged to room temperature (Fig. 3). In Fig. 4, the ratio $\Delta R/R=[R(H)-R(H=0)]/R(H=0)$ versus H is plotted for 5, 50, 145, and 300 K. The ratio $\Delta R/R$ changes from 25% at 5 K to 15% at 300 K. This effect is comparatively much larger than the negative MR, which is observed in the case of the InSb substrate at low magnetic fields and low temperatures as part of an otherwise intense positive magnetoresistance background (inset, Fig. 4). In the case of the bulk InSb the small negative MR is similar to the weak disorder effect observed in *n*-type InSb by Mani *et al.*[13] It is interesting to mention that in older literature the negative MR observed at low magnetic fields and low temperatures in *n*-type degenerate semiconductors is attributed to the scattering of the conduction electrons by localized spins through an

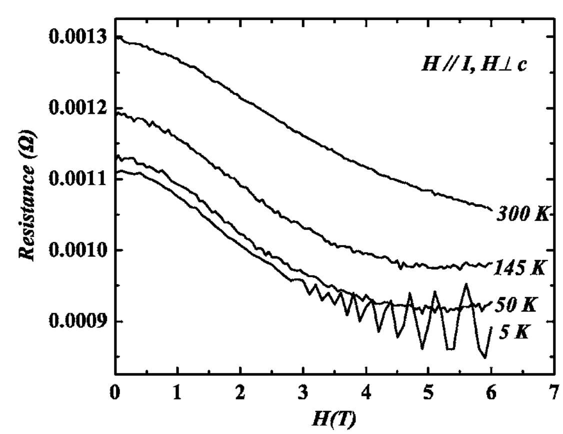

FIG. 3. Plots of resistance as a function of the magnetic field (H) at 5, 50, 145, and 300 K. H is applied in plane ($H\perp c$) and parallel to the current direction ($H\parallel I$).

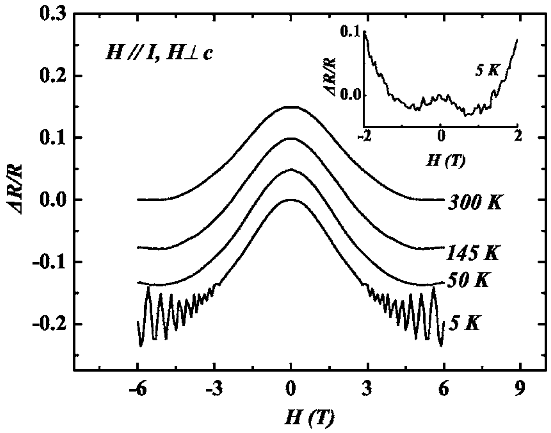

FIG. 4. Plots of the ratio $\Delta R/R$ as a function of the magnetic field (H) at 5, 50, 145, and 300 K. H is applied in plane ($H \perp c$) parallel to the current direction ($H \parallel I$). The curves are shifted along the y axis for clarity. Inset: Plot of the ratio $\Delta R/R$ versus H at 5 K for the InSb substrate with the same H configuration. (The low-magnetic-field region is shown in order to highlight the small negative MR.)

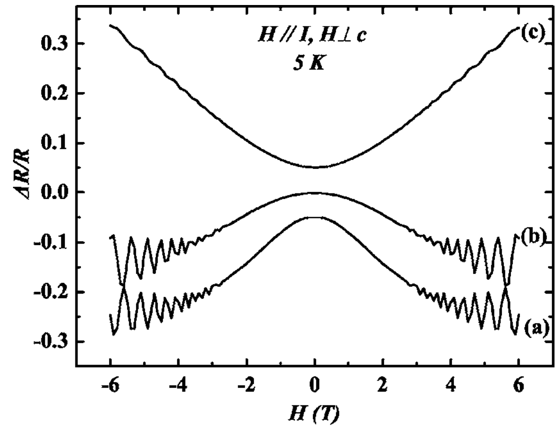

FIG. 5. Plots of the ratio $\Delta R/R$ as a function of the magnetic field (H) for (a) NiMnSb/InSb, (b) Ni/InSb, and (c) Al/InSb at 5 K. H is applied in plane ($H \perp c$) and parallel to the current direction ($H \parallel I$). The curves are shifted along the y axis for clarity.

s-d exchange interaction although magnetic impurities are not present.[14–17]

It is noteworthy that negative GMR effect is observed neither in NiMnSb bulk samples nor in NiMnSb films deposited on highly resistive Si substrates[18] (in this case we measure only the magnetoresistance of the NiMnSb films) with the same H configuration. In addition, no negative GMR is observed for the configuration in which H is in plane but perpendicular to the current [Fig. 2(b)].

To investigate the origin of this interesting negative longitudinal GMR effect, we performed similar MR measurements on Ni/InSb and Al/InSb systems for which Ni and Al films were deposited on heated InSb substrates (200 °C) by PLD. Whereas we observed a negative GMR in the longitudinal MR of the magnetic Ni/InSb system, in the case of the nonmagnetic Al/InSb system we observed only a positive MR without any negative MR component (Fig. 5). Moreover, we measured the longitudinal MR of a NiMnSb/InSb system, where the NiMnSb film was deposited by PLD at room temperature [Fig. 6(a)]. In this case only a small negative MR was found on an otherwise positive MR background. Interestingly, after annealing the room-temperature-grown sample in a pure Ar atmosphere at 200 °C for 30 min, the negative MR increased drastically [Fig. 6(b)]. In all of these systems we observed SdH oscillations in the MR at 5 K and their *I-V* characteristic curves exhibited Ohmic behavior.

Upon measuring the MR of the metal/InSb systems, we observe the contribution of both the metallic film and the InSb substrate because they are both conductive. This is evident at low temperatures, where we observe the SdH oscillations originating from the substrate. In addition, the fact that the negative GMR is not observed in any of the constituents of the NiMnSb/InSb or Ni/InSb systems leads us to suggest that this effect occurs at the interface between the ferromagnetic metal and InSb. A plausible explanation for the negative GMR is that at the interface there is a thin layer of InSb containing microscopic magnetic entities (e.g., NiMnSb, Ni precipitates). Upon increasing the magnetic field, these magnetic entities gradually align their magnetic moments with the external magnetic field leading to a decrease in the spin-dependent resistance of the system. These precipitates are ablated metal particles that reach the surface of the InSb substrate with energies high enough to allow their implantation and penetration to a shallow depth.

To substantiate this conjecture we have actually done XRR measurements on 5- and 37-nm-thin NiMnSb/InSb films. Figure 7 shows the scattering length density profile of these two NiMnSb/InSb films as function of film thickness. In both of the films, there is a distinct low-density layer (15% less than the bulk density of the film) between InSb substrate and the film. The thickness of this low-density layer is 3.5 nm in both samples. This is different from what has been observed earlier for similar thickness of NiMnSb films on Si substrates.[10] In this system, there exists a 3–5 nm low-

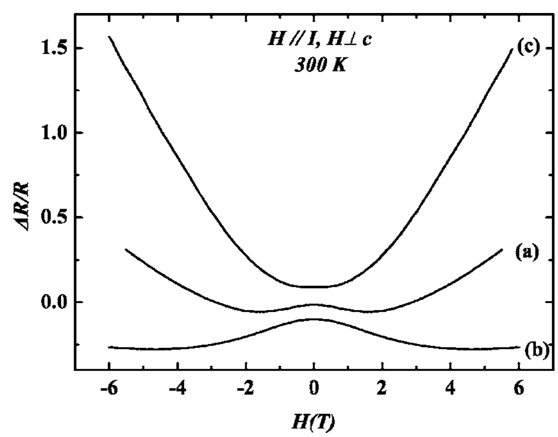

FIG. 6. Plots of the ratio $\Delta R/R$ versus H at 300 K for the NiMnSb/InSb system: (a) the NiMnSb film is deposited by PLD at room temperature, (b) following postdeposition annealing at 200 °C in Ar, and (c) the NiMnSb film is deposited at 200 °C at a pressure of 0.05 mbar of Ar. H is applied in plane ($H \perp c$) and parallel to the current direction ($H \parallel I$). The curves are shifted along the y axis for clarity.

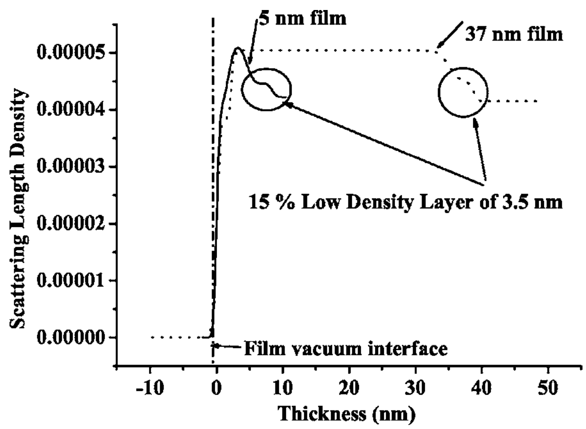

FIG. 7. Scattering length density profile of NiMnSb thin films (5 and 35 nm) on InSb substrate as a function of film thickness.

density NiMnSb layer and additionally a 3–4 nm SiO_2 layer below it. As a result we observed no negative MR at the NiMnSb/Si interface. Both of the following reasons could be responsible for this observation. (a) Due to the presence of the SiO_2 layer the magnetic entities cannot penetrate deeply enough to reach the surface of the Si substrate. (b) The SiO_2 layer is an insulating layer. Moreover, the Si substrate had a higher resistivity than InSb (InSb exhibited metallic behavior). Thus, most of the current should flow through the NiMnSb film in contrast to the case of the NiMnSb/InSb system in which we expect the current to flow also through the interface and the InSb substrate as we have argued earlier. On the other hand, there was a distinct formation of porous layer on those NiMnSb/Si films, which is not observed in the present NiMnSb/InSb films.

It has been reported that either reduction of the laser fluence and/or increase of the ambient pressure can slow down the energetic particles during PLD deposition.[19] To this end, we introduced pure Ar gas to a pressure of 0.05 mbar in order to reduce the velocities of the ablated particles. Figure 6(c) indicates clearly that there is no negative MR in this case. Thereby our data indicate that this implantationlike process depends on the energy of the ablated particles and the thermal processing of the system. Specifically, once the particles are energetic enough the temperature could promote clustering of the particles, or assist diffusion deeper into InSb, or assist in recovering the high mobility at the interface, or, finally, a combination of two or more of the above events could occur.

A more striking evidence of the possible existence of spin scatterers at the interface in the NiMnSb/InSb and Ni/InSb systems is that the negative GMR effect does not occur in the Al/InSb system because Al precipitates in InSb have no magnetic moment and therefore, only the positive magnetoresistance contribution is observed. It is worth mentioning that the observed negative GMR is similar to the one observed in Cu-Co alloys consisting of ultrafine Co precipitates in a Cu matrix when the magnetic field is applied parallel to the current.[5,6] It is also similar to the negative MR effects observed in GaAs films containing MnAs ferromagnetic clusters[7] and in Mn_xGe_{1-x} dilute ferromagnetic semiconductors containing MnGe ferromagnetic clusters.[8] It is also noteworthy that negative MR has been observed in erbium-, yttrium-, and europium-doped InSb films and has been attributed to scattering of conduction electrons by the magnetic spins of the aforementioned rare earth atoms.[20–22] In all of these cases, however, the negative GMR effect does not persist at room temperature. On the contrary, in the system under investigation, the negative GMR effect does not depend strongly on temperature and remains large even at room temperature. Finally, the quadratic dependence of the negative GMR on the magnetic field observed in our samples for magnetic fields up to 2 T is in agreement with theoretical calculations of negative MR due to localized spins.[23] The strong MR anisotropy of the investigated system which depends on the direction of the current flow through the magnetic precipitates relatively to the external applied magnetic field, as shown in Fig. 2, could be utilized for sensing moderate to high magnetic fields.

IV. CONCLUSION

In summary, we have investigated the MR properties of the NiMnSb/InSb system fabricated by depositing NiMnSb thin films on InSb substrates by PLD. An unusual negative GMR effect persisting at room temperature is observed when the magnetic field is applied in plane and parallel to the current. This effect is attributed to magnetic precipitates formed at the interface between the ferromagnetic metal film and the InSb substrate. This argument was substantiated by XRR measurements which revealed the presence of a low-density layer of NiMnSb at the interface between NiMnSb and InSb. The strong MR anisotropy of the system could have interesting magnetic sensor applications.

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