

# Measurement of spin polarization by Andreev reflection in ferromagnetic $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ epilayers

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We carried out point contact Andreev reflection (PCAR) spin spectroscopy measurements on epitaxially grown ferromagnetic  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  epilayers with a Curie temperature of  $\sim 9$  K. The spin sensitivity of PCAR in this material was demonstrated by parallel control studies on its nonmagnetic analog,  $\text{In}_{1-y}\text{Be}_y\text{Sb}$ . We found the conductance curves of the Sn point contacts with  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  to be fairly conventional, with the possible presence of proximity-induced superconductivity effects at the lowest temperatures. These measurements provided control data for subsequent PCAR measurements on ferromagnetic  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ , which indicated spin polarization in  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  to be  $52 \pm 3\%$ . © 2004 American Institute of Physics. [DOI: 10.1063/1.1760883]

The recent emergence of III–Mn–V dilute ferromagnetic semiconductor alloys, such as  $\text{In}_{1-x}\text{Mn}_x\text{As}$  (Ref. 1) and  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ,<sup>2</sup> has already led to a number of exciting results relevant to spintronics applications. The ability to fabricate tunneling magnetoresistance (TMR) devices<sup>3</sup> naturally integrated with technologically important semiconductors, such as GaAs, makes these materials especially attractive. The need to increase operating temperature has stimulated extensive studies of  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  epilayers, where the highest Curie temperature is now close to 190 K. While the ferromagnetic  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  alloy<sup>4–6</sup> is much less explored, it has—in spite of its lower Curie temperature—significant potential for application in infrared *spin photonics* and in spin transport devices due to its lighter holes, small energy gap, and much higher carrier mobility than in other III–Mn–V ferromagnetic semiconductors.

The efficiency of most of spintronic devices, such as giant magnetoresistance (GMR) or TMR junctions, depends on the *transport* spin polarization of carriers in ferromagnetic materials. Point Contact Andreev Reflection (PCAR)<sup>7,8</sup> has recently been introduced as an effective technique for measuring the transport spin polarization  $P_c$  of metals and metallic oxides<sup>9</sup> in the ballistic limit,

$$P_c = \frac{\langle N_{\uparrow}(E_f) v_{f\uparrow} \rangle - \langle N_{\downarrow}(E_f) v_{f\downarrow} \rangle}{\langle N_{\uparrow}(E_f) v_{f\uparrow} \rangle + \langle N_{\downarrow}(E_f) v_{f\downarrow} \rangle},$$

where  $N_{\uparrow}(E_f)$  and  $N_{\downarrow}(E_f)$ ,  $v_{f\uparrow}$  and  $v_{f\downarrow}$  are the densities of states and the Fermi velocities for majority and minority spin subbands, respectively. PCAR is based on the difference in the Andreev reflection process in normal metal/superconductor (N/S) and in ferromagnet/superconductor (F/S) contacts.<sup>10</sup> While at the clean N/S interface all quasiparticles with the energies  $eV \leq \Delta$  ( $\Delta$  is the superconductor

energy gap) are converted into Cooper pairs, at the F/S interface uncompensated quasiparticles are unable to propagate, thus reducing the conductance and affecting the overall character of the conductance curves,  $dI/dV$ , which can then be related to the degree of spin polarization of the ferromagnet.<sup>11</sup>

The key to extending the PCAR technique to the FSm/S interface is the requirement of *high junction transparency*, which is often limited by native Schottky barriers present at most semiconductor/superconductor (Sm/S) interfaces. Most of the experimental work on Andreev reflection in semiconductors, starting with the pioneering work of Kastalsky *et al.*,<sup>12</sup> has been done in a two-dimensional (2D) configuration. While the Andreev process for metallic F/S interfaces, in spite of some remaining theoretical questions,<sup>13</sup> is fairly well established, the Sm/S interface is relatively unexplored<sup>14</sup> and a number of important theoretical questions still remain unanswered. In particular, a complete theory of Andreev reflection in magnetic semiconductors, which should include the effects of spin–orbit interaction, magnetic and structural disorder, is yet to be developed.

In our recent work,<sup>15</sup> we have demonstrated conventional three-dimensional (3D) Andreev reflection in a nonmagnetic semiconductor using low temperature (LT)-GaAs doped up to a very high level with Be, that produced a free hole concentration as high as  $p = 8 \times 10^{20} \text{ cm}^{-3}$ . These measurements suggested, that PCAR spectroscopy could be also successfully applied to dilute magnetic semiconductors, at least to those with high carrier concentrations. However, initial Andreev reflection studies on FSm/S junctions using  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  epilayers with carrier concentrations comparable to that of our  $\text{Ga}_{1-y}\text{Be}_y\text{As}$ , carried out both by us<sup>15</sup> and by a Florida State University group,<sup>16</sup> indicated unconventional behavior, possibly arising from some not yet understood interaction between the superconductor and the mag-

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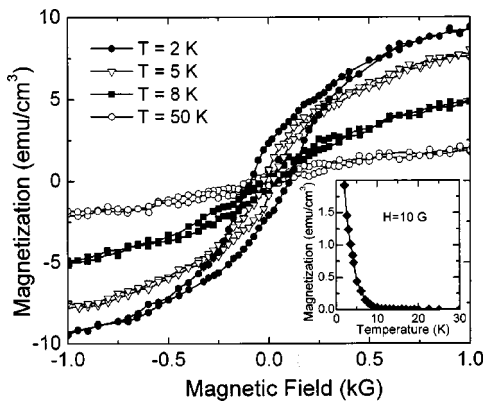


FIG. 1. Field dependence of the magnetization in  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  epitaxial film with  $x=0.028$ . The data were collected for a series of temperatures with the field applied perpendicular to the layer plane. The inset shows the temperature dependence of the remanent magnetization, indicating Curie temperature of  $\sim 9$  K.

netic semiconductor. These results suggest that interpretation of PCAR experiments on ferromagnetic semiconductors could be facilitated by direct comparison with Andreev reflection measurements carried out on analogous nonmagnetic semiconductors. Additionally, to separate different mechanisms contributing to observed effects, it is important to extend Andreev reflection studies to other members of the III–Mn–V family of ferromagnetic semiconductors, especially those with very different physical parameters from  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ . In this respect  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ , with its effective mass the lowest among III–Vs, and hence the highest in mobility, is an excellent candidate, since it is expected to form high-transparency FSm/S junctions. Moreover, it is fortunate that its nonmagnetic analog,  $\text{In}_{1-y}\text{Be}_y\text{Sb}$ , needed for comparative studies, can also be grown by low temperature molecular beam epitaxy.

In this letter we report PCAR spin polarization measurements of  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  epilayers with Curie temperature of  $\sim 9$  K. In order to facilitate interpretation of the  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  results, these measurements are accompanied by a detailed comparison with the Andreev reflection data obtained on its nonmagnetic analog,  $\text{In}_{1-y}\text{Be}_y\text{Sb}$ .

The growth of both  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  and  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  films was carried out by LT-molecular beam epitaxy, described in detail in Refs. 4 and 5. The In and Mn (Be) fluxes were supplied from standard effusion cells, while  $\text{Sb}_2$  flux was produced by a Sb cracker cell. Hybrid (100) CdTe/GaAs and InSb/AlSb/GaAs substrates were used for growth. Prior to depositing each  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  or  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  film we grew a 100 nm LT-InSb buffer layer at  $210^\circ\text{C}$  which was seen from a well-resolved reflection high energy electron diffraction (RHEED) pattern, provided a flat surface for subsequent deposition. The substrate was then cooled to  $170^\circ\text{C}$  for growth of a 230-nm-thick LT- $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  or LT- $\text{In}_{1-y}\text{Be}_y\text{Sb}$  epilayer. A 3:1  $\text{Sb}_2$ :In beam equivalent pressure ratio was used for the growth of both InSb-based systems.

We studied several  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  epilayers with  $x \approx 0.03$ . The specimens had mobility of  $\sim 150$   $\text{cm}^2/\text{sV}$  and a Curie temperature of approximately 9 K, as determined from the temperature dependence of the remanent magnetization [see Fig. 1], and from the series of hysteresis curves taken at different temperatures [Fig. 1]. For the nonmagnetic analog

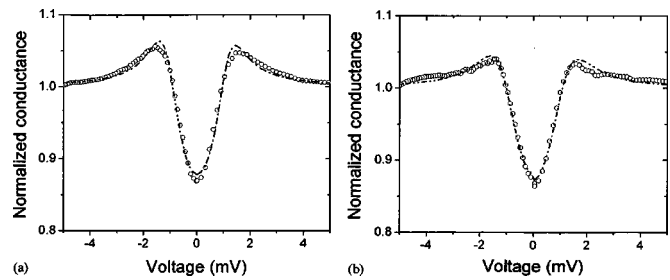


FIG. 2. Typical normalized conductance curves for two different Sn superconductor contacts with  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  epitaxial films: (a) contact resistance  $R_c = 57 \Omega$ ,  $T = 1.2$  K;  $\Delta = 0.52$  mV; fitting parameters:  $Z = 0.19$  and  $P = 54\%$ ; (b) contact resistance  $R_c = 36 \Omega$ ,  $T = 1.6$  K;  $\Delta = 0.5$  mV; fitting parameters:  $Z = 0.20$  and  $P = 52\%$ .

to be used as the control sample we chose an epilayer of  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  with  $y = 0.05$ , selected in such a way that its free hole concentration  $p = 1.5 \times 10^{20} \text{ cm}^{-3}$  was close to that of the ferromagnetic film ( $p \sim 2 \times 10^{20} \text{ cm}^{-3}$ ; see Ref. 6).

Mechanically sharpened Sn superconductor tips were used for all the point contact measurements reported in this study. The conductance was measured by the standard four-terminal technique using lock-in detection at 2 kHz. Conductance curves were analyzed by means of a modified BTK model<sup>11</sup> with two fitting parameters: the spin polarization  $P_c$  and the dimensionless interfacial scattering parameter  $Z$ . Details of the measurement and fitting procedures are given in Refs. 17 and 11, respectively.

In contrast to  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ,<sup>15</sup> all measurements of  $\text{Sn}/\text{In}_{1-x}\text{Mn}_x\text{Sb}$  point contacts indicate fairly conventional Andreev reflection behavior of a ferromagnet/superconductor junction. Representative  $dI/dV$  curves for two different contacts at 1.2 and 1.6 K are shown in Fig. 2. The superconductor gap value used for analysis of all our data,  $\Delta(0) \sim 0.52$  mV, was close to the bulk value for Sn. The gap at higher temperatures was obtained from the Bardeen–Cooper–Schrieffer (BCS)  $\Delta(T)$  dependence, with the critical temperature of Sn close to its bulk value,  $T_c \sim 3.7$  K. All of the experimental  $dI/dV$  curves display a characteristic dip at zero bias, which is consistent with the suppression of Andreev reflection due to the spin polarization of  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ .

The value of spin polarization  $P_c$  for  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  was obtained after analyzing each of the  $dI/dV$  curves for every contact at corresponding temperatures using the theory given in Ref. 11. The average spin polarization was found to be  $P_c \sim 52 \pm 3\%$ . The typical values of  $Z \leq 0.25$  indicate high junction transparency. It is important to note that even with nominally no barrier contact some quasiparticle reflection is always present due to the so-called Fermi velocity mismatch  $r = v_s/v_{\text{Sm}}$  between a superconductor and a semiconductor,<sup>14</sup> where  $v_s$  and  $v_{\text{Sm}}$  are the respective Fermi velocities. To evaluate this Fermi velocity mismatch in both the  $\text{Sn}/\text{In}_{1-x}\text{Mn}_x\text{Sb}$  and  $\text{Sn}/\text{In}_{1-x}\text{Be}_x\text{Sb}$  contacts, we used the density-of-states effective mass in InSb (Ref. 18),  $m^* = 0.25 m_0$  and the free hole concentration  $p = 2 \times 10^{20} \text{ cm}^{-3}$  to estimate the Fermi velocities in InSb. Taking the value of the Fermi velocity of Sn to be  $1.9 \times 10^8$  cm/s, we obtain  $r = 2.3$ , which yields a minimum  $Z$  value of  $\sim 0.4$ . These estimates are in good agreement with the experimental  $Z$  values obtained for  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  and for  $\text{In}_{1-x}\text{Be}_x\text{Sb}$  as described below.

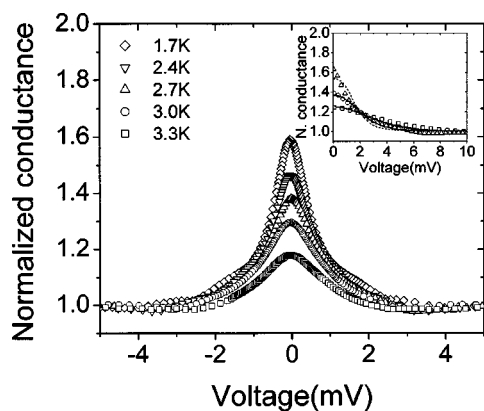


FIG. 3. Normalized conductance as a function of the voltage for a Sn superconductor contact with an  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  epitaxial film (contact resistance of  $R_c=63\ \Omega$ ) for a series of temperatures. The inset shows fitting of the experimental data (solid curve) for three different temperatures. The  $Z$  parameter for all the fitted curves is  $0.39\pm 0.05$ .

From the known values of the hole density and the resistivity,  $\rho\sim 0.2\ \text{m}\Omega\ \text{cm}$ , we can estimate the low temperature mean free path  $L$  for light and heavy holes to be  $\sim 60$  and  $\sim 15\ \text{nm}$ , respectively. The contact size  $d$  can be estimated from the Sharvin formula,  $R_n=(4\rho L/3\pi d^2+\rho/2d)\times(1+Z^2)$ , where  $R_n$  is the contact resistance. For typical values of the contact resistance  $R_n\sim 60\ \Omega$ , we have obtained contact size  $d\sim 25\ \text{nm}$ , indicating that our measurements were done in the ballistic regime,  $L\geq d$ .

To test the reliability of our measurements in the spin-polarized  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  system, we have studied a large number of nonmagnetic  $\text{Sn}/\text{In}_{1-x}\text{Be}_x\text{Sb}$  junctions. A series of characteristic conductance curves for one of these contacts at different temperatures is shown in Fig. 3. In contrast to the  $\text{Sn}/\text{In}_{1-x}\text{Mn}_x\text{Sb}$  contacts, these curves exhibit higher conductance at zero bias compared to their normal conductance above the gap. This behavior, together with the increase of zero bias conductance at lower temperatures, is consistent with Andreev reflection for high-transparency junctions. All fitted curves (see inset in Fig. 3) have a *temperature-independent* value of approximately  $0.39\pm 0.05$ , in good agreement with the  $Z$  values obtained for the analogous  $\text{InMnSb}$  system, as well as with the estimates based on the Fermi velocity mismatch obtained above. At lower temperatures, however, we consistently observe some discrepancy between the experimental curves and the best fit, as can be seen from the upper curve in the inset of Fig. 3. This discrepancy, which is especially pronounced in some of the lowest-resistance contacts, may be due to the presence of proximity-induced superconductivity<sup>19</sup> in the  $\text{In}_{1-x}\text{Be}_x\text{Sb}$  film.

In summary, we have measured transport spin polarization of the dilute ferromagnetic semiconductor  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ . Simultaneously, we made a detailed study of the analogous system  $\text{In}_{1-y}\text{Be}_y\text{Sb}$ , which served as a nonmagnetic control material for PCAR measurements on  $\text{InMnSb}$ . The measurements in both magnetic and non-magnetic systems demonstrate fairly conventional Andreev reflection in high-transparency junctions, as the interface scattering parameter,  $Z$  is close to the minimum values estimated from the Fermi

velocity mismatch. We have not observed any measurable density of states broadening and/or superconducting gap suppression. The spin polarization of  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  was determined to be  $52\pm 3\%$ . The measurements on  $\text{In}_{1-y}\text{Be}_y\text{Sb}$  indicate a possible influence of the proximity effect on the junction properties. While the behavior of the  $\text{Sn}/\text{In}_{1-y}\text{Be}_y\text{Sb}$  contacts is similar to the behavior of the  $\text{Sn}/\text{Ga}_{1-y}\text{Be}_y\text{As}$  system,<sup>15</sup> there is a marked difference between the behavior of Sn contacts with  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  and with  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ . Narrower band gap as well as a much higher carrier mobility, characteristic of  $\text{In}_{1-x}\text{Mn}_x\text{Sb}$  compared to  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ , may be just several of a number of important parameters that affect the physics of Andreev reflection in these systems, and it is our hope that this experimental study will stimulate the development of the theory of Andreev reflection for III–Mn–V ferromagnetic semiconductors.

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- <sup>1</sup>H. Munekata, H. Ohno, S. von Molnar, A. Segmüller, L. L. Chang, and L. Esaki, *Phys. Rev. Lett.* **63**, 1849 (1989).
- <sup>2</sup>H. Ohno, A. Shen, F. Matsukura, A. Oiwa, A. Endo, S. Katsumoto, and Y. Iye, *Appl. Phys. Lett.* **69**, 363 (1996).
- <sup>3</sup>M. Tanaka and Y. Higo, *Phys. Rev. Lett.* **87**, 026602 (2001); M. S. H. Chun, S. J. Potashnik, K. C. Ku, P. Schiffer, and N. Samarth, *Phys. Rev. B* **66**, 100408 (2002).
- <sup>4</sup>T. Wojtowicz, G. Cywinski, W. L. Lim, X. Liu, M. Dobrowolska, J. K. Furdyna, K. M. Yu, W. Walukiewicz, G. B. Kim, M. Cheon, X. Chen, S. M. Wang, and H. Luo, *Appl. Phys. Lett.* **82**, 4310 (2003).
- <sup>5</sup>T. Wojtowicz, W. L. Lim, X. Liu, G. Cywinski, M. Kutrowski, L. V. Titova, K. Yee, M. Dobrowolska, J. K. Furdyna, K. M. Yu, W. Walukiewicz, G. B. Kim, M. Cheon, X. Chen, S. M. Wang, H. Luo, I. Vurgaftman, and J. R. Meyer, *Physica E (Amsterdam)* **20**, 325 (2004).
- <sup>6</sup>S. Yanagi, K. Kuga, T. Slupinski, and H. Munekata, *Physica E (Amsterdam)* **20**, 333 (2004).
- <sup>7</sup>R. J. Soulen, Jr., J. M. Byers, M. S. Osofsky, B. Nadgorny, T. Ambrose, S. F. Cheng, P. R. Broussard, C. T. Tanaka, J. Nowak, J. S. Moodera, A. Barry, and J. M. D. Coey, *Science* **282**, 85 (1998).
- <sup>8</sup>S. K. Upadhyay, A. Palanisami, R. N. Louie, and R. A. Buhrman, *Phys. Rev. Lett.* **81**, 3247 (1998).
- <sup>9</sup>I. Zutic, J. Fabian, and S. Das Sarma, *Rev. Mod. Phys.* (in press).
- <sup>10</sup>M. J. M. de Jong and C. W. J. Beenakker, *Phys. Rev. Lett.* **74**, 1657 (1995).
- <sup>11</sup>I. I. Mazin, A. A. Golubov, and B. Nadgorny, *J. Appl. Phys.* **89**, 7576 (2001).
- <sup>12</sup>A. Kastalsky, A. W. Kleinsasser, L. H. Greene, R. Bhat, F. T. Milliken, and J. P. Harbison, *Phys. Rev. Lett.* **67**, 3026 (1991).
- <sup>13</sup>K. Xia, P. J. Kelly, G. E. W. Bauer, and I. Turek, *Phys. Rev. Lett.* **89**, 166603 (2002).
- <sup>14</sup>I. Zutic and S. Das Sarma, *Phys. Rev. B* **60**, R16322 (1999).
- <sup>15</sup>R. P. Panguluri, K. C. Ku, N. Samarth, T. Wojtowicz, X. Liu, J. K. Furdyna, Y. Lyanda-Geller, and B. Nadgorny (unpublished).
- <sup>16</sup>J. G. Braden, J. S. Parker, P. Xiong, S. H. Chun, and N. Samarth, *Phys. Rev. Lett.* **91**, 056602 (2003).
- <sup>17</sup>R. P. Panguluri, G. Tsoi, B. Nadgorny, S. H. Chun, N. Samarth, and I. I. Mazin, *Phys. Rev. B* **68**, 201307(R) (2003).
- <sup>18</sup>I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, *J. Appl. Phys.* **89**, 5815 (2001).
- <sup>19</sup>S. Han, L. F. Cohen, and E. L. Wolf, *Phys. Rev. B* **42**, 8682 (1990); A. Kadin, *ibid.* **41**, 4070 (1990).