

# Large Spin Accumulation Signals in Epitaxial $\text{Mn}_5\text{Ge}_3$ Contacts on Ge without Oxide Tunnel Barrier

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## Abstract

**Spin injection in high-quality epitaxial  $\text{Mn}_5\text{Ge}_3$  Schottky contacts on n-type Ge has been investigated using a three-terminal Hanle effect measurement. Clear Hanle and inverted Hanle signals with features characteristic of spin accumulation and spin precession are observed up to 200 K. Strikingly, the observed spin voltage is several orders of magnitude larger than predicted by the theory of spin injection and diffusive spin transport. Since the devices have no oxide tunnel barrier, the discrepancy between theory and experiments cannot be explained by the often-invoked spin accumulation in localized states associated with the oxide or oxide/semiconductor interface. The observed spin voltages therefore must originate from the Ge itself, either from spins in the Ge bulk bands or its depletion region.**

## 1. Introduction

Great progress in the electrical spin injection and detection in semiconductors has reinforced the field of spin-based semiconductors. To achieve efficient spin injection into a semiconductor (SC), magnetic tunnel contacts consisting of a conventional 3d-ferromagnet (FM) and an oxide tunnel barrier have been widely used [1-8]. Subsequently, the presence of spin accumulation in the SC can be detected by electrical means using either a local three-terminal (3T) geometry [1] or a non-local (NL) four-terminal geometry [9].

Recently, the origin of the spin accumulation signals observed with the 3T configuration has been heavily debated. Experimental data obtained by many research groups [1,2,4-8] are many orders of magnitude larger than what the theory for spin injection and diffusion predicts [10]. To explain the large spin voltages, the first and most often-cited theory involves two-step tunneling through localized states near the oxide/SC interface [2,11]. While recent results indicate that this theoretical explanation is not correct [8], the topic is still under discussion. Thus, it was considered that the spin signal may originate from the oxide tunnel barrier itself [2,7,12-14], possibly from localized states in the oxide close the interface with the ferromagnet or from localized states deep within the tunnel oxide. More recently, the original theory of two-step tunneling via localized states has been extended by including the on-site Coulomb repulsion [13,14]. We stress that all these explanations [2,7,11-14] involve localized states, either in the tunnel oxide or at the

oxide/semiconductor interface, and also the existence of spin accumulation and precession.

Understanding the novel underlying physics of spin transport in 3T devices is therefore an important goal, and the answer cannot be found in NL devices where the large signal enhancement is generally not observed. Considering this, we focus on 3T devices, and investigate another approach to create a spin accumulation in a SC by tunneling, namely via a FM/SC Schottky tunnel contact. This has been done previously using specific FM alloys such as  $\text{Fe}_3\text{Si}$  [15]. A direct Schottky contact is an interesting system for 3T spin transport because there is no oxide tunnel barrier. This eliminates all possible enhancements of the spin voltage by localized states in the tunnel oxide or at the oxide/SC interface. Nevertheless, we show here that in such Schottky contacts without oxide tunnel barrier, the spin voltage is also much larger than predicted by theory.

## 2. Fabrication of the Schottky devices

The epitaxial  $\text{Mn}_5\text{Ge}_3$  Schottky contact was fabricated on an As-doped Ge(111) substrate with a carrier concentration of  $1.1 \times 10^{18} \text{ cm}^{-3}$  and a resistivity of 70 m $\Omega\text{cm}$  at 300 K. To form the  $\text{Mn}_5\text{Ge}_3$  compound, we carried out the solid phase epitaxy method [16] by first depositing a 8 nm-thick Mn layer at room temperature (RT), and then annealing the sample in-situ at 450  $^\circ\text{C}$ . The transmission electron microscopy (TEM) image presented in Fig. 1(a) confirmed that an epitaxial  $\text{Mn}_5\text{Ge}_3$  film with a high crystalline quality was obtained. However, in the low magnification TEM image (Fig. 1(b)), a non-negligible roughness is visible. For

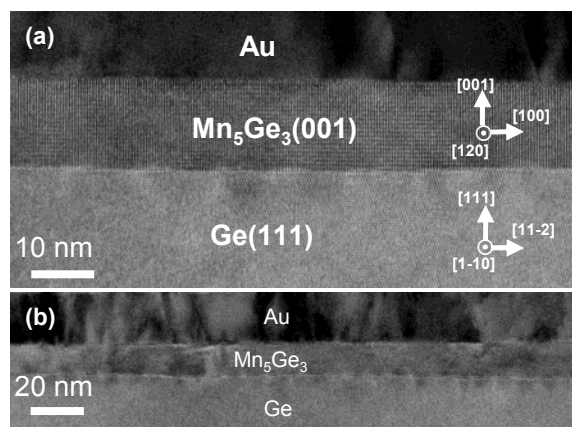


Fig. 1 (a) High-resolution and (b) low magnification cross-sectional TEM image of the Au/ $\text{Mn}_5\text{Ge}_3$ /n-Ge structure.

the transport measurements, standard micro-fabrication techniques were carried out to fabricate the Schottky junctions with an active area of  $100 \times 200 \mu\text{m}^2$ .

### 3. Results and discussion

To probe the presence of spin accumulation, we performed 3T Hanle effect measurements [1]. In this geometry the FM Schottky contact acts as a spin injector and detector whereas two other contacts are used as the reference electrodes. For a normal Hanle measurement, a magnetic field ( $B_\perp$ ) perpendicular to the film plane and thus transverse to the spins reduces the spin accumulation due to spin precession, inducing a change in the voltage across the junction, at constant current. In addition, the roughness of the FM can create local stray fields that induce a partial depolarization of the spins at zero magnetic field. This can be suppressed by applying an in-plane magnetic field ( $B_\parallel$ ), known as inverted Hanle effect [17]. Fig. 2(a) and (b) presents the change in the voltage across the  $\text{Mn}_5\text{Ge}_3/\text{n-Ge}$  Schottky contact measured at 15 and 200 K under forward bias and at a constant current. The spin voltage obtained show all the characteristic features of precession of an induced non-equilibrium spin population: a signal decay with a Lorentzian shape for small magnetic fields perpendicular to the spins, a signal recovery for larger perpendicular fields due to rotation of the ferromagnetic injector into the direction of the applied field, and an inverted Hanle effect for applied magnetic fields parallel to the spin direction. This also allows us to rule out tunneling anisotropic magnetoresistance, anisotropic magnetoresistance and Hall voltages since these do not produce a signal with a Lorentzian line shape.

We compare the experimental value of the spin- $RA$  to the value predicted from the standard theory for spin injection in a nonmagnetic material [10]. The theoretical spin- $RA$  is equal to  $P^2 \rho_{\text{Ge}} l_{\text{sd}}$  where  $P$  is the tunnel spin polarization of  $\text{Mn}_5\text{Ge}_3$ ,  $\rho_{\text{Ge}}$  the resistivity of the Ge substrate and  $l_{\text{sd}}$  the spin-diffusion length. Taking  $P \sim 0.5$  and  $\rho_{\text{Ge}} = 0.07 \Omega\text{cm}$  at 15 K, and assuming  $l_{\text{sd}} = 1 \mu\text{m}$ , we obtain a theoretical spin- $RA$  of  $175 \Omega\mu\text{m}^2$ . The measured spin- $RA$  is  $100 \text{ k}\Omega\mu\text{m}^2$  at 15 K, which is about 3 orders of magnitude larger than the theoretical value. Moreover, the spin voltage is comparable to that previously observed for FM/oxide/Ge tunnel junctions [4,5,8].

As previously mentioned, it has frequently been argued that the enhanced Hanle spin signals originate from localized states at the oxide/SC interface or originate from the oxide barrier itself [2,7,12-14]. However, since the Schottky contacts studied here do not have an oxide tunnel barrier, the enhanced spin signal cannot be explained by any mechanism that involves the oxide and the localized states these may produce. Moreover, our Ge substrate has a homogeneous doping density, so that we can avoid complications due to presence of a potential well or a  $\delta$ -doping layer that arise when the semiconductor surface is heavily doped, as in previous work [15,18]. We note that interface states, i.e., metal-induced gap states, are certainly expected to be

present in FM/SC Schottky contacts. However, such states are directly coupled to the FM and hence these interface states cannot sustain a large spin accumulation as spins will be drained away by the FM that acts as an efficient spin sink. Therefore, we conclude that the large spin signal can only come from spins in the Ge itself, either from spins in the Ge bulk bands or its depletion region.

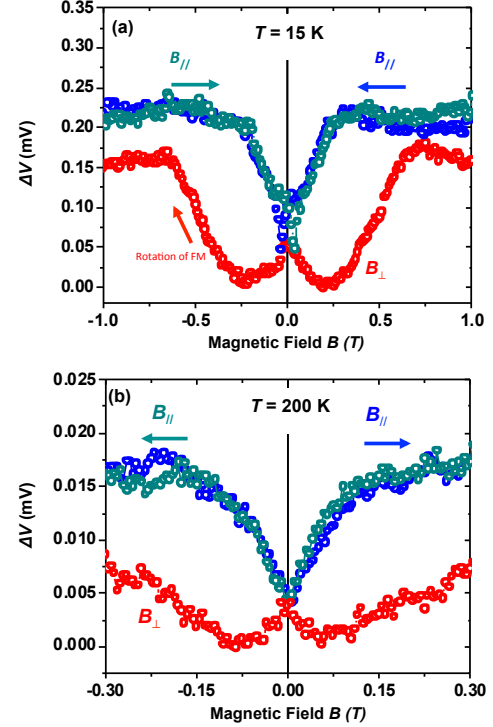


Fig. 2 Hanle ( $B_\perp$ ) and inverted Hanle ( $B_\parallel$ ) curves of the  $\text{Mn}_5\text{Ge}_3/\text{n-Ge}$  device measured at (a) 15 K with  $V \sim -300 \text{ mV}$  and  $I = -40 \mu\text{A}$  and (b) 200 K with  $V \sim -300 \text{ mV}$  and  $I = -3.37 \text{ mA}$  (electron extraction from the Ge into the FM).

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### References

- [1] S. P. Dash *et al.*, Nature **462**, 491 (2009).
- [2] M. Tran *et al.*, Phys. Rev. Lett. **102**, 036601 (2009).
- [3] T. Suzuki *et al.*, Appl. Phys. Express **4**, 023003 (2011).
- [4] K.-R. Jeon *et al.*, Phys. Rev. B **84**, 165315 (2011).
- [5] S. Iba *et al.*, Appl. Phys. Express **5**, 053004 (2012).
- [6] M. Ishikawa *et al.*, Appl. Phys. Lett. **100**, 252404 (2012).
- [7] T. Uemura *et al.*, Appl. Phys. Lett. **101**, 132411 (2012).
- [8] S. Sharma *et al.*, Phys. Rev. B **89**, 075301 (2014).
- [9] F. J. Jedema *et al.*, Nature **410**, 345 (2001).
- [10] A. Fert and H. Jaffrès, Phys. Rev. B **64**, 184420 (2001).
- [11] R. Jansen *et al.*, Phys. Rev. B **85**, 134420 (2012).
- [12] O. Txoperena *et al.*, Appl. Phys. Lett. **102**, 192406 (2013).
- [13] Y. Song *et al.*, arXiv:1401.7649 (2014).
- [14] O. Txoperena *et al.*, arXiv:1404.0633v1 (2014).
- [15] Y. Ando *et al.*, Appl. Phys. Lett. **94**, 182105 (2009).
- [16] C. Zeng *et al.*, Appl. Phys. Lett. **83**, 5002 (2003).
- [17] S. P. Dash *et al.*, Phys. Rev. B **84**, 054410 (2011).
- [18] Q. O. Hu *et al.*, Phys. Rev. B **84**, 085306 (2011).