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1           **Spin injection and detection up to room temperature in**  
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13                           (Dated: December 6, 2016)

14                           **Abstract**

15           We have measured the spin injection efficiency and spin lifetime in Co<sub>2</sub>FeSi/*n*-GaAs lateral  
16 nonlocal spin valves from 20 to 300 K. We observe large ( $\sim 40 \mu\text{V}$ ) spin valve signals at room  
17 temperature and injector currents of  $10^3 \text{ A/cm}^2$ , facilitated by fabricating spin valve separations  
18 smaller than the  $1 \mu\text{m}$  spin diffusion length and applying a forward bias to the detector contact.  
19 The spin transport parameters are measured by comparing the injector-detector contact separation  
20 dependence of the spin valve signal with a numerical model accounting for spin drift and diffusion.  
21 The apparent suppression of the spin injection efficiency at the lowest temperatures reflects a  
22 breakdown of the ordinary drift-diffusion model in the regime of large spin accumulation. A  
23 theoretical calculation of the D'yakonov-Perel spin lifetime agrees well with the measured *n*-GaAs  
24 spin lifetime over the entire temperature range.

## 25 I. INTRODUCTION

26 All-electrical spin transport has been demonstrated in III-V semiconductors [1–4], group  
27 IV semiconductors [5], and in 2D materials such as graphene [6, 7]. One of the most mature  
28 systems studied in the field of semiconductor spintronics is the ferromagnet (FM)/*n*-GaAs  
29 lateral spin valve (SV) structure [1–3]. GaAs-based devices have served as a testbed for  
30 several seminal semiconductor (SC) spin transport measurements, such as the Hanle effect [1,  
31 8], the spin Hall and inverse spin Hall effects [9–11], and nuclear hyperfine effects [8, 12–14].  
32 The Dresselhaus spin-orbit interaction (SOI) [15] originating from the non-centrosymmetric  
33 lattice of III-V SCs makes them attractive candidates for modulation of spin transport using  
34 the SOI [16]. At the same time, however, the Dresselhaus SOI present in III-V SCs leads to  
35 efficient spin relaxation in the diffusive transport regime.

36 Electron spin relaxation in *n*-GaAs at doping levels near the metal-insulator transition  
37 is governed by the D'yakonov-Perel (DP) mechanism [17, 18]. The DP spin relaxation rate  
38 in III-V semiconductors has a characteristic  $\tau_s^{-1} \propto \epsilon^3$  behavior [17, 19], where  $\epsilon$  is the  
39 carrier energy. The spin lifetime  $\tau_s$  is the inverse of the spin relaxation rate. At tempera-  
40 tures for which the carriers are nondegenerate ( $\epsilon \sim k_b T$ ), the spin lifetime falls sharply as  
41  $\tau_s \propto T^{-3}$  [20]. Short spin lifetimes ( $\sim 10 - 100$  ps) have therefore challenged *n*-GaAs SV  
42 room temperature performance [4], as the short spin lifetime limits the steady-state spin  
43 accumulation.

44 In this article we demonstrate electrical detection of nonlocal spin accumulation in Heusler  
45 alloy FM/*n*-GaAs lateral spin valve devices up to room temperature. Clear nonlocal SV  
46 signals are measured by fabricating devices with injector-detector contact separations of less  
47 than a spin diffusion length and applying a forward bias voltage to the detector contact.  
48 We use the injector-detector contact separation dependence of the SV signal to extract  
49 the *n*-GaAs spin lifetime and FM/SC interface spin injection efficiency from 20 K up to  
50 room temperature. These data allow for a comprehensive and quantitative evaluation of  
51 the temperature-dependent performance of FM/*n*-GaAs lateral SV devices. We find that  
52 the spin lifetime in the *n*-GaAs channel is in quantitative agreement with a theoretical  
53 calculation of the DP spin lifetime over the entire temperature range. At low temperatures,  
54 we achieve a spin accumulation that is a significant fraction of the carrier density in the  
55 channel. This is accompanied by an apparent downturn in the injection efficiency which

56 we believe is due to breakdown of the ordinary drift-diffusion model in the regime of large  
57 spin-dependent electrochemical potential splitting.

## 58 II. METHODS

### 59 A. Structure growth and device fabrication

60 The devices used in this study were fabricated from heterostructures grown by molecular-  
61 beam epitaxy (MBE). A 2.5  $\mu\text{m}$  Si-doped ( $n = 3 \times 10^{16} \text{ cm}^{-3}$ ) GaAs epilayer was grown  
62 following a 500 nm undoped GaAs buffer layer grown on a semi-insulating (001) GaAs sub-  
63 strate. To thin the naturally occurring Schottky depletion layer and provide a tunnel barrier  
64 for efficient spin injection [21–23], the doping level was increased at the FM/SC interface. A  
65 15 nm transitional doping layer was grown ( $n = 3 \times 10^{16} \text{ cm}^{-3} \rightarrow n^+ = 5 \times 10^{18} \text{ cm}^{-3}$ ) on top  
66 of the  $n$ -GaAs epilayer, followed by an 18 nm thick heavily doped ( $n^+ = 5 \times 10^{18} \text{ cm}^{-3}$ ) layer.  
67 Following the GaAs MBE growth, the sample was cooled to  $< 400^\circ \text{ C}$  under  $\text{As}_4$ -flux at which  
68 point the  $\text{As}_4$ -flux was turned off. This resulted in a highly ordered GaAs(001) $c(4 \times 4)$  As-rich  
69 surface reconstruction as confirmed by reflection high-energy electron diffraction (RHEED)  
70 and *in situ* scanning tunneling microscopy (STM). For the 5 nm thick epitaxial Heusler  
71 film growth, the samples were transferred to a separate growth chamber while maintaining  
72 ultra-high vacuum (UHV). The Heusler film growth was performed at  $270^\circ \text{ C}$  with codepo-  
73 sition from individual elemental sources. The Heusler compounds grow with a cube-on-cube  
74 orientation with Heusler(001) $\langle 110 \rangle \parallel$  GaAs(001) $\langle 110 \rangle$ [24, 25]. During Heusler growth  
75 RHEED was used to confirm layer-by-layer growth of a single crystal film. Cross-sectional  
76 high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM)  
77 was performed, and example images of the interfaces are shown in Fig. 1. These images  
78 confirm the samples are single crystals with mixed  $L2_1$  and B2 phases in both  $\text{Co}_2\text{MnSi}$   
79 (Fig. 1(a)) and  $\text{Co}_2\text{FeSi}$  (Fig. 1(b)) films, and a degree of intermixing at the GaAs/Heusler  
80 interface of no more than 4-6 atomic layers. The GaAs(001)/Heusler interface resulted in a  
81 uniaxial magnetic anisotropy yielding an easy axis along the GaAs [110] direction [24, 26, 27]  
82 for both the  $\text{Co}_2\text{FeSi}$  and  $\text{Co}_2\text{MnSi}$  films.

83 The heterostructures were patterned into lateral spin valve devices using a top-down  
84 fabrication process. A combination of electron-beam lithography and photolithography was

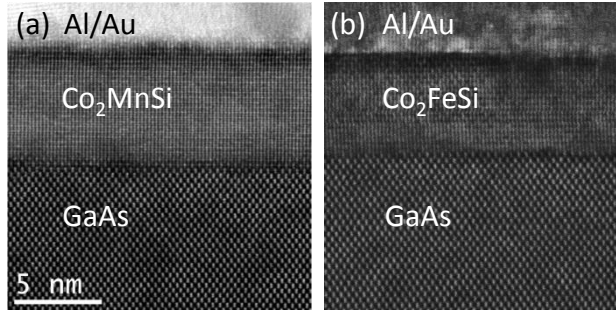


FIG. 1. Cross-sectional HAADF-STEM images of (a) the  $\text{Co}_2\text{MnSi}/\text{GaAs}$  interface and (b) the  $\text{Co}_2\text{FeSi}/\text{GaAs}$  interface. Images (a) and (b) were taken on the same heterostructures used for the  $\text{Co}_2\text{MnSi}$  and  $\text{Co}_2\text{FeSi}$  spin valves measurements presented in this paper. A 5 nm scale bar is indicated in the lower left of (a).

85 used, with  $\text{Ar}^+$  ion milling to define the ferromagnetic contacts and wet etching to define  
 86 the  $n$ -GaAs channel. A silicon nitride insulating layer was deposited by plasma-enhanced  
 87 chemical vapor deposition (PECVD) and patterned by lift-off to electrically isolate the  
 88 evaporated Ti/Au vias and bonding pads from the substrate and  $n$ -GaAs channel sidewalls.  
 89 A micrograph of a SV device is shown in Fig. 2(a). The channel width in the GaAs [110]  
 90 direction is  $80\ \mu\text{m}$ , the SV contact length is  $50\ \mu\text{m}$ , the injector width is  $1\ \mu\text{m}$ , and the  
 91 detector width is  $0.5\ \mu\text{m}$ . The large aspect ratio of the SV contacts along the magnetic  
 92 easy axis was chosen in order to minimize fringe magnetic fields as well as to define a two-  
 93 dimensional geometry conducive to modeling (channel width  $\gg$  spin diffusion length). The  
 94 large-area remote contacts share the same composition as the SV contacts. The remote  
 95 contacts, however, have no impact on the SV measurement, because they are placed many  
 96 spin diffusion lengths away from the SV contacts. Multiple SV devices were fabricated on  
 97 the same chip by wet etching through the  $2.5\ \mu\text{m}$   $n$ -GaAs to isolate the devices electrically.  
 98 SV devices on the same chip were patterned with injector-detector edge-to-edge separations  
 99 ranging from 250 nm to  $5\ \mu\text{m}$ .

## 100 B. Charge transport

101 Standard multiprobe dc transport measurements were performed as a function of tem-  
 102 perature to characterize both the  $n$ -GaAs channel and the  $\text{Co}_2\text{FeSi}/n$ -GaAs interface. A

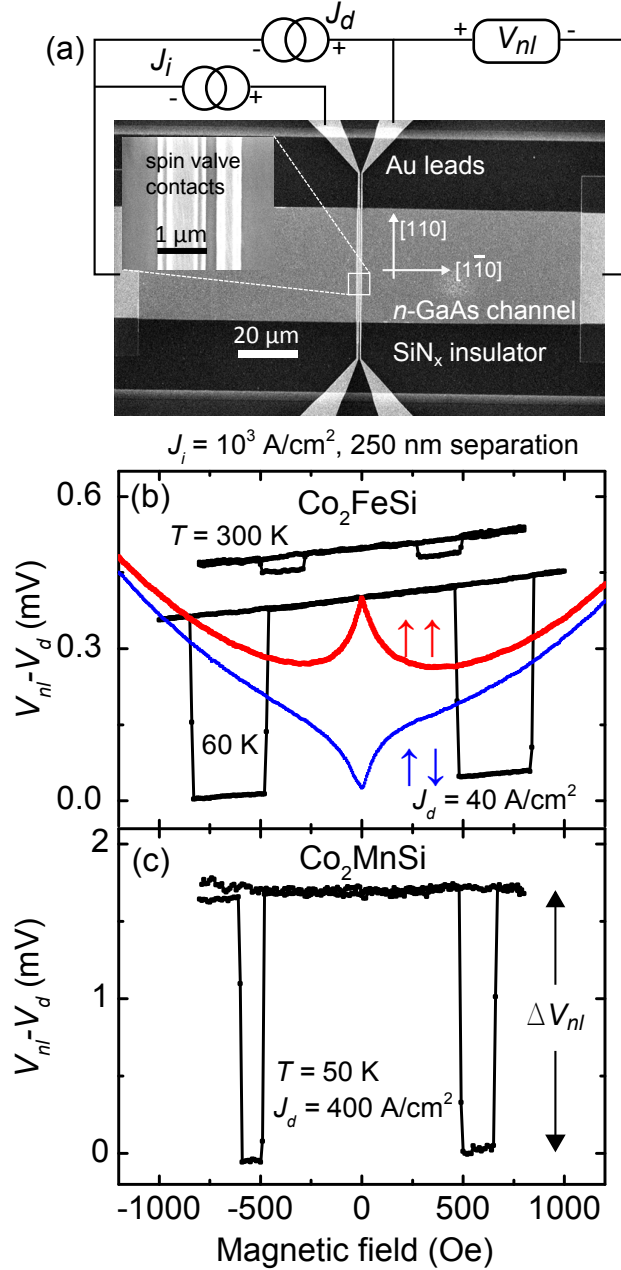


FIG. 2. (Color online) (a) Scanning electron micrograph of a lateral SV device, with a schematic diagram of the measurement. The inset is a magnified image of the injector (left contact) and detector (right contact), in the device pictured with an edge-to-edge separation of 250 nm. (b-c) Example BDSV field sweeps for devices with Co<sub>2</sub>FeSi contacts (b) and Co<sub>2</sub>MnSi contacts (c). The temperature and bias conditions are indicated on the figure.  $\Delta V_{nl}$  is the magnitude of the parallel-antiparallel difference as indicated in (c). At the bias conditions indicated in (b)  $V_d = 0.44 \text{ V}$  at 60 K and  $V_d = 0.30 \text{ V}$  at 300 K. In (c)  $V_d = 0.72 \text{ V}$  at 50 K for the bias conditions indicated. After subtracting  $V_d$ , the 60 K and 300 K data in (b) are offset for clarity. In (b), the dc NLH measurement is shown at 60 K, for both parallel (red) and antiparallel (blue) magnetization configurations.

103 companion Hall bar was fabricated from the same heterostructure used to fabricate the SV  
 104 devices, and transport measurements were performed from 10-350 K to extract the carrier  
 105 concentration and mobility of the  $n$ -GaAs. The Hall carrier concentration was measured to  
 106 be  $2.8 \times 10^{16} \text{ cm}^{-3}$  for the  $\text{Co}_2\text{FeSi}$  heterostructure and  $3.5 \times 10^{16} \text{ cm}^{-3}$  for the  $\text{Co}_2\text{MnSi}$   
 107 heterostructure. Fig. 3(a) shows the channel electron mobility and diffusion constant as a  
 108 function of temperature for the  $\text{Co}_2\text{FeSi}$  heterostructure. The Hall factor [28], which causes  
 109 deviation of the Hall mobility from the electron mobility in  $n$ -GaAs, is accounted for by  
 110 assuming the Hall factor is unity at 300 K [29, 30] and that the carrier concentration is  
 111 temperature-independent.

112 A typical SV device  $\text{Co}_2\text{FeSi}/n$ -GaAs contact three-terminal (3T) interface current-  
 113 voltage ( $J - V$ ) characteristic is shown in Fig. 3(b). The inset of Fig. 3(b) shows the  
 114 differential conductance per unit area ( $dJ/dV$ ) as a function of temperature. Tunneling-  
 115 dominated transport (field emission) is known to be necessary for spin injection in FM/GaAs  
 116 Schottky contacts [31]. The existence of tunneling-dominated transport under forward bias  
 117 at all temperatures is supported by two observations. First,  $dJ/dV$  increases exponentially  
 118 with forward bias voltage at all temperatures, at a rate that is independent of tempera-  
 119 ture. Because of the triangular Schottky barrier [32], the forward bias voltage across a  
 120 Schottky interface changes the thickness of the effective potential barrier through which  
 121 tunneling occurs [33, 34]. Although thermionic emission and thermionic field emission also  
 122 lead to an exponential increase of  $dJ/dV$  with interface forward bias voltage, the rate for  
 123 those processes is strongly temperature-dependent, ruling out those mechanisms. Second,  
 124 at temperatures below the Fermi temperature of the  $n$ -GaAs ( $\sim 60$  K for these samples)  
 125 the forward bias differential conductance decreases weakly with decreasing temperature. Al-  
 126 though  $dJ/dV$  at forward bias is temperature-dependent above the Fermi temperature, this  
 127 does not imply thermionic emission but rather an increase in the tunneling attempt rate  
 128 due to the nondegeneracy of the  $n$ -GaAs [33].

### 129 C. Spin transport

130 A schematic diagram of the SV measurement is shown in Fig. 2(a). A dc bias current  $J_i$   
 131 flows through the injector contact and a second bias current  $J_d$  flows through the detector  
 132 contact. The injector and detector current sources share a common remote reference contact.

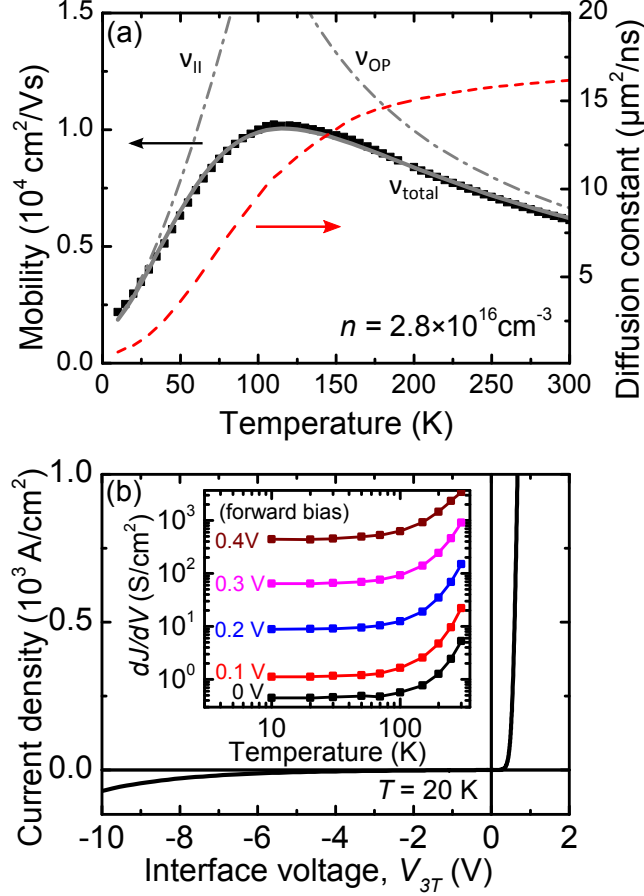


FIG. 3. (Color online) (a) The  $n$ -GaAs mobility extracted from Hall measurements (left ordinate) as a function of temperature on the  $\text{Co}_2\text{FeSi}$  heterostructure. The gray solid line is a fit to the model for the mobility given by Eq. 10, with the ionized-impurity (II) and optical-phonon (OP) scattering contributions to the mobility indicated with the dash-dot gray lines. In the fit shown,  $A = 1.3 \times 10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ ,  $B = 18 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\text{K}^{-3/2}$  and  $C = 2.0 \times 10^6 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\text{K}^{-1}$ . The red dashed line (right ordinate) is the channel diffusion constant calculated with Eq. 6. (b) Typical  $\text{Co}_2\text{FeSi}$  contact 3-terminal  $J - V$  characteristic at 20 K. The inset in (b) is the differential conductance as a function of temperature at different interface forward bias voltages. The solid curves connect data points.

133 In this article positive currents and interface voltages refer to electron extraction from the  
 134 channel, *i.e.*, forward bias of the metal/semiconductor Schottky contact. The bias current  
 135 applied to the detector contact results in a voltage drop  $V_d$  over the tunnel barrier, which is  
 136 the 3T interface voltage of the detector contact. In these devices, a forward bias applied at  
 137 the detector contact enhances the nonlocal SV signal size compared to an unbiased detector



138 (zero detector bias is the traditional nonlocal SV configuration pioneered by Johnson and  
 139 Silsbee [35]). We will henceforth refer to the case of a bias current applied through the  
 140 detector contact as the biased-detector spin valve (BDSV) measurement. The enhancement  
 141 in the SV signal size with a bias applied to the detector contact has been observed in prior  
 142 *n*-GaAs lateral SV literature on similar heterostructures [36, 37], and the possible origins  
 143 will be discussed in detail later in this article.

144 An applied magnetic field is swept along the FM easy axis to switch the magnetizations  
 145 of the injector and detector contacts from the parallel to antiparallel configuration, which  
 146 allows for a definitive measurement of the nonlocal voltage due to spin accumulation. The  
 147 difference in the nonlocal detector voltage  $V_{nl}$  between the parallel and antiparallel contact  
 148 magnetization states is due to spin accumulation in the semiconductor [35] and is given by

$$\Delta V_{nl} = V_{NL,\uparrow\uparrow} - V_{NL,\uparrow\downarrow} = \eta(V_d) \frac{n_{\uparrow} - n_{\downarrow}}{e} \frac{\partial\mu}{\partial n}, \quad (1)$$

149 where  $n_{\uparrow(\downarrow)}$  is the majority (minority) spin-resolved carrier density in the GaAs channel,  
 150  $e$  is the electron charge, and  $\partial\mu/\partial n$  is the inverse of the thermodynamic compressibility  
 151 of the semiconductor. We will refer to  $n_{\uparrow} - n_{\downarrow}$  as the spin accumulation and  $(n_{\uparrow} - n_{\downarrow})/n$   
 152 as the dimensionless spin polarization throughout this article. The dimensionless detection  
 153 efficiency parameter  $\eta(V_d)$  characterizes the spin sensitivity of the detection contact [38] and  
 154 is a function of the bias voltage. Because of the bias current applied through the detector  
 155 contact,  $V_{nl}$  is not an open circuit nonlocal voltage (or “electromotive force”). The voltage  
 156 drop over the detector Schottky tunnel barrier contributes an offset  $V_d$ , so that

$$V_{nl} = V_d + \frac{\Delta V_{nl}}{2} \hat{\mathbf{m}}_i \cdot \hat{\mathbf{m}}_d \quad (2)$$

157 where  $\hat{\mathbf{m}}_{i(d)}$  is the unit vector specifying the magnetization of the injector (detector) contact.

158 Example BDSV field sweeps are shown in Figs. 2(b) and (c) on SV devices with an  
 159 injector-detector edge-to-edge separation of 250 nm at an injector bias current of  $J_i = 10^3$   
 160 A/cm<sup>2</sup>. The BDSV measurement on the device with Co<sub>2</sub>FeSi contacts is shown in Fig. 2(b)  
 161 at  $J_d = 40$  A/cm<sup>2</sup>, and for the device with Co<sub>2</sub>MnSi contacts in Fig. 2(c) at  $J_d = 400$   
 162 A/cm<sup>2</sup>. The Co<sub>2</sub>MnSi/*n*-GaAs contacts exhibited large voltage noise in the nonlocal SV  
 163 measurements, and the signal-to-noise ratio (SNR) was not adequate for measurements at  
 164 high temperatures. For this reason, the analysis presented in this article is carried out  
 165 for measurements on Co<sub>2</sub>FeSi/*n*-GaAs devices. At low temperatures, at which the SNR in

166 Co<sub>2</sub>MnSi/*n*-GaAs devices was adequate, the SV measurements were quantitatively similar  
167 to those on Co<sub>2</sub>FeSi/*n*-GaAs devices. A linear background in  $V_{nl}$  can result from the Hall  
168 effect due to slight misalignment. The slope, which is a weak function of temperature, is  
169 subtracted from the data before extracting  $\Delta V_{nl}$ .

170 Nonlocal Hanle (NLH) measurements [35, 39] were also performed in the biased-detector  
171 configuration. In the NLH measurement a magnetic field applied perpendicular to the  
172 sample plane is used to apply a precessional torque, which, in combination with diffusion,  
173 dephases the spin accumulation. In all of the NLH measurements, the applied field was  
174 small enough so that the out-of-plane rotation of the contact magnetization decreased the  
175 in-plane component of the magnetization by less than 1.5%, which was considered negligible.  
176 The NLH measurement could be executed with the injector and detector contacts in either  
177 the parallel or antiparallel configuration. In the fitting of the NLH lineshape discussed in  
178 Section III D, the difference of the parallel and antiparallel field sweeps is used.

179 At cryogenic temperatures, the NLH measurement in *n*-GaAs is complicated by the  
180 strong hyperfine fields due to dynamic nuclear polarization (DNP) [12, 14, 19]. Steady-state  
181 conditions are difficult to achieve due to long ( $\sim$  seconds) nuclear depolarization timescales,  
182 and small misalignments between the applied field and the contact magnetization result in  
183 oblique Overhauser fields, which distort the NLH lineshape [12, 14]. To mitigate the influence  
184 of DNP effective fields on the NLH lineshape, a low duty cycle ( $< 1\%$ ) pulsed current  
185 measurement was used for the NLH sweeps at temperatures below 100 K. The current was  
186 turned off for 1000 milliseconds, then pulsed on for 5 milliseconds after which the voltage  
187 was recorded and the pulse-train repeated. The current rise and fall times were much shorter  
188 than the few-millisecond current pulse duration. The pulsed measurement minimizes the  
189 nuclear polarization buildup because the current is on for a time much less than the nuclear  
190 polarization time [19]. Example NLH data obtained for the 250 nm separation Co<sub>2</sub>FeSi  
191 device at 60 K are shown in Fig. 2(b).

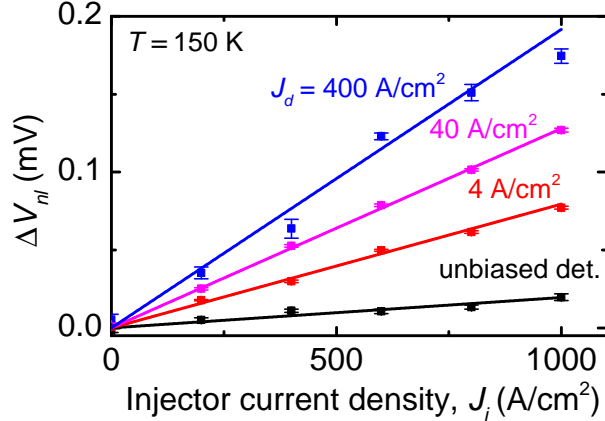


FIG. 4. (Color online) Injector bias current dependence of  $\Delta V_{nl}$ , for varying detector forward bias currents, on the 250 nm separation device at 150 K. The lines shown are linear fits.

### 192 III. RESULTS

#### 193 A. Effect of detector bias

194 We now discuss the effect of detector bias on our SV measurements. First, we note that  
 195 Crooker *et al.* [36] and Bruski *et al.* [37] observed similar enhancement of the spin valve  
 196 signal in the presence of a detector bias current or voltage. Although several mechanisms  
 197 have been proposed to explain the enhancement in the nonlocal SV signal with detector  
 198 bias, the enhancement remains poorly understood. At the end of this section, we will return  
 199 to discuss possible explanations in light of our measurements.

200 We find that a sufficiently large forward bias current applied through the detector contact  
 201 increases the SV signal  $\Delta V_{nl}$  at all temperatures. Fig. 4 shows  $\Delta V_{nl}$  vs.  $J_i$  for the 250 nm  
 202 separation at 150 K.  $\Delta V_{nl}$  increases linearly with  $J_i$  at all detector bias currents, but the slope  
 203 of  $\Delta V_{nl}$  vs.  $J_i$  is enhanced with increasing detector forward bias current. This enhancement  
 204 is particularly advantageous for measurements at high temperatures near 300 K, at which  
 205 the spin valve signal becomes small in *n*-GaAs [1, 4]. This effect was observed in devices  
 206 with both Co<sub>2</sub>FeSi and Co<sub>2</sub>MnSi contacts and was observed previously for devices with Fe  
 207 contacts [36].  
 208

209 For the case of no bias current passing through the detector (*i.e.* the conventional nonlocal  
 210 SV measurement),  $\Delta V_{nl}$  could be measured in the 250 nm separation device for temperatures

211 less than approximately 200 K (see data points in Fig. 5(b-c) at  $V_d = J_d = 0$ ). For a fixed  
 212 injector current, the SV measurement was then performed at different detector bias currents.  
 213 The corresponding interface voltage drop  $V_d$  was measured at each bias current, and so the  
 214 data may be presented as a function of either bias voltage  $V_d$  or current  $J_d$ . The results  
 215 of this measurement at 60 K on the 250 nm separation are shown in Fig. 5(a) and are  
 216 summarized for all temperatures in Figs. 5(b) and (c). At forward detector bias above  
 217 interface voltages of  $V_d \sim 0.2$  V, we observe significant enhancement of  $\Delta V_{nl}$ . As shown in  
 218 Fig. 5(a), the dependence of  $\Delta V_{nl}$  on the detector bias is non-monotonic below  $\sim 200$  K, and  
 219 it is suppressed at small detector voltages (of either sign) and even changes sign for a narrow  
 220 window of reverse bias. Although  $V_{nl}$  is sensitive to 3T signals [8] produced by *local* spin  
 221 injection at the detector contact, only *nonlocally*-injected spin accumulation contributes  
 222 to  $\Delta V_{nl}$  in a spin valve measurement, because  $\Delta V_{nl}$  is the difference in nonlocal voltage  
 223 between parallel and antiparallel magnetization states. Furthermore, as shown in Fig. 2(b),  
 224 the NLH measurement can also be performed with the parallel-antiparallel difference at zero  
 225 field matching the BDSV magnitude. The existence of the NLH effect at low temperatures  
 226 demonstrates conclusively that the biased-detector measurement in these devices is a probe  
 227 of the nonlocally injected spin accumulation.

229 The enhancement in  $\Delta V_{nl}$  under forward detector bias occurs at all temperatures mea-  
 230 sured, from 20 K to room temperature. Using the BDSV measurement a clear SV signal  
 231 could be measured on the separations below 1  $\mu\text{m}$  up to and above room temperature on the  
 232  $\text{Co}_2\text{FeSi}$  devices. To our knowledge, the spin signal we measure on the 250 nm separation  
 233 device of  $\sim 40$   $\mu\text{V}$  at room temperature is over an order-of-magnitude larger than that which  
 234 has been achieved in FM/*n*-GaAs SVs, to date [4]. We now discuss the possible origins of  
 235 the forward bias enhancement of the SV signal.

236 We consider first the influence of drift due to electric fields in the channel between the  
 237 injector and detector contacts. Due to the relatively low carrier density in these samples,  
 238 the spin drift length  $l = \tau_s J / ne$  can be comparable to or larger than the spin diffusion  
 239 length  $\lambda = \sqrt{D\tau_s}$  [40, 41]. In the case of a forward bias current applied through the detector  
 240 contact (electron extraction from the channel), the electric field in the channel causes drift  
 241 of electrons from the injector towards the detector contact, enhancing the nonlocal spin  
 242 accumulation when compared to spin diffusion alone. To determine if the detector bias  
 243 current leads to significant drift enhancement of  $\Delta V_{nl}$ , the current density in the channel

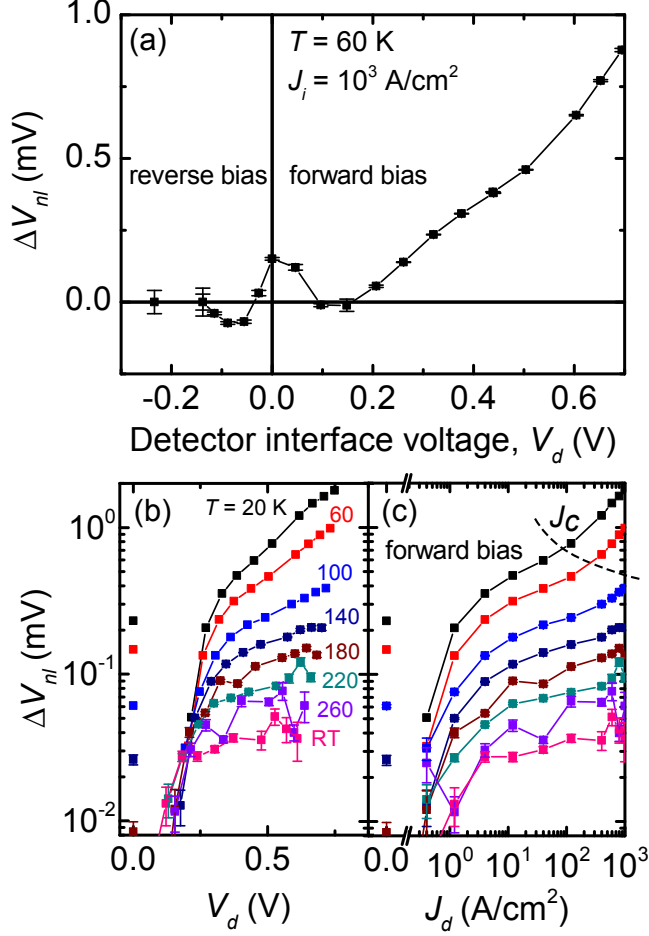


FIG. 5. (Color online) (a)  $\Delta V_{nl}$  as a function of detector interface voltage  $V_d$  for fixed injector bias current. (b,c) The detector forward bias voltage (b) and current (c) dependence of  $\Delta V_{nl}$  from 20 K to room temperature (RT). Only the zero detector bias and forward bias points are shown in (b) and (c) to illustrate the enhancement of  $\Delta V_{nl}$  at forward detector bias. The dashed line in (c) indicates  $J_c$ , above which spin drift in the channel caused by the detector bias current enhances the spin accumulation at the detector. For clarity, the dashed line was drawn to smoothly connect  $J_c$  at each temperature. All data shown in this figure were taken with the 250 nm injector-detector separation device, and  $J_i = 10^3$  A/cm<sup>2</sup>.

244 between injector and detector contacts at which the spin drift length was equal to the  
 245 spin diffusion length was evaluated at each temperature. Above a critical current density  
 246  $J_c = ne\sqrt{D/\tau_s}$ , which is the current density at which  $l = \lambda$ , drift enhancement of the  
 247 nonlocal spin accumulation below the detector contact becomes significant. The region  
 248 where this occurs is illustrated in Fig. 5(c), in which the dashed curve shows  $J_c$ . The

249 drift enhancement is significant only at low temperatures and the highest detector bias  
 250 currents. This is in contrast to the case of Si described in Ref. [41] in which the long spin  
 251 lifetime at room temperature, combined with higher current densities than we apply, leads  
 252 a spin drift length which can be much longer than the spin diffusion length. Because the  
 253 enhancement in  $\Delta V_{nl}$  occurs at all temperatures and for current densities far below  $J_c$ , it  
 254 cannot be attributed solely to spin drift effects in the channel. Although variations on simple  
 255 drift models have been proposed [42], it is unlikely that drift alone can play a significant  
 256 role given that the enhancement is observed up to room temperature. For the purposes of  
 257 discussion, we attribute the enhancement in  $\Delta V_{nl}$  with detector forward bias primarily to  
 258 enhancement of  $\eta$ , the detection efficiency, which we treat as a purely interfacial property.  
 259 The detection efficiency is a function of detector bias, *i.e.*  $\eta \rightarrow \eta(V_d)$ .

260 Hu *et al.* [43] and Salis *et al.* [3] observed a highly non-monotonic behavior of the sign  
 261 of the injected spin polarization in similar heterostructures with Fe contacts. The sign and  
 262 magnitude depended strongly on the details of the  $n$ -GaAs band structure in the region of  
 263  $n^+$  doping near the interface. It is possible that the enhancement of  $\eta$  under forward bias  
 264 is due to the enhanced participation of additional quantum well states that form on the SC  
 265 side of the tunnel barrier due to the  $n^+$  doping layer. It has been proposed that these states  
 266 play a critical role in both charge and spin current in tunnel contacts using Schottky barriers  
 267 through FM/SC wavevector-matching arguments which depend on the degree of quantum  
 268 confinement of the SC states [44].

269 Another point of view focuses on the nonlinear current-voltage characteristic of the tunnel  
 270 barrier itself [45, 46]. A simple analysis suggests that the ratio of the detected voltage to  
 271 the spin accumulation should be modified by the ratio  $(J/V)/(dJ/dV)$  of the absolute to  
 272 differential conductance, although Jansen *et al.* [47] have noted that this correction factor is  
 273 in fact an upper bound. In our case, however, we observe an effect that is opposite to that  
 274 suggested by this argument.  $(J/V)/(dJ/dV)$  is smaller at forward bias voltage than at zero  
 275 bias, because  $J$  increases exponentially with  $V$ .

276 Because the bias current applied to the detector introduces a 3T offset  $V_d$  to  $V_{nl}$ , care  
 277 must be taken to separate signals due to nonlocal spin accumulation from signals of local  
 278 origin. Surface localized states in tunnel barriers have been at the center of a controversy  
 279 in the semiconductor spin injection literature because of the influence these states can have  
 280 on both the magnitude and lineshape of the 3T Hanle measurement [48]. For example,

281 Txoperena *et al.* [49] determined that impurity-assisted tunnelling processes can lead to  
 282 Lorentzian-shaped magnetoresistance effects that mimic the Hanle effect. Also, Jansen *et al.*  
 283 [50] note that in the 3T geometry the change in 3T voltage due to spin accumulation can  
 284 originate from spin accumulation in interface localized states as well as bulk channel spin  
 285 accumulation. Our measurement, however, probes the parallel-antiparallel difference in  
 286 the nonlocal voltage, notwithstanding the bias applied to the detector contact. Although  
 287 localized states may play an important role in the spin-polarized transport at our interfaces,  
 288 the mechanisms discussed by Txoperena *et al.* [49], Jansen *et al.* [50] are only relevant for 3T  
 289 local spin detection where the ferromagnetic contact simultaneously serves as the injector  
 290 and detector.

291 Another possible physical explanation for the detector bias dependence of  $\Delta V_{nl}$  is that  
 292 significant features exist in the spin-resolved density-of-states (DOS) of the  $\text{Co}_2\text{FeSi}/\text{GaAs}$   
 293 interface near the Fermi level. These features could lead to spin injection and detection  
 294 efficiencies that vary with forward bias voltage, as states above the Fermi level in the FM  
 295 become available for elastic tunnelling from the SC. Density functional theory (DFT) calcula-  
 296 tions done for  $\text{Co}_2\text{FeSi}$  in the  $\text{L2}_1$  phase[51, 52] suggest strong variations in the bulk minority  
 297 DOS near the Fermi level over energy ranges of  $\sim$ hundreds of meV, which are comparable to  
 298 the scale of the interface voltages at the detector in our measurement. Strong bulk minority  
 299 DOS variations near the Fermi level have also been predicted for  $\text{Co}_2\text{MnSi}$  which are largely  
 300 insensitive to the phase ( $\text{L2}_1$  vs.  $\text{B2}$ )[53]. However, the bias dependence of spin detection  
 301 shown in Fig. 5(a) cannot be clearly correlated with the features in the spin-resolved DOS  
 302 reported by DFT calculations. Additionally, interface states, such as those which have been  
 303 proposed for the  $\text{Fe}/\text{GaAs}(001)$  interface, will contribute to the tunneling current[54]. Al-  
 304 though it is likely that the low-voltage features in  $\Delta V_{nl}(V_d)$  are associated with electronic  
 305 structure of the interface, we have no quantitative description of the bias-dependence of the  
 306 nonlocal voltage.

307 We now comment briefly on the sign of the spin valve signals we observe. In this article,  
 308 a decrease in  $V_{nl}$  in the antiparallel magnetization state is defined as a positive  $\Delta V_{nl}$ . The  
 309 BDSV sweeps shown in Figs. 2(b) and (c) are examples of positive  $\Delta V_{nl}$  values. The sign  
 310 of  $\Delta V_{nl}$  is determined by the relative signs of the injection and detection efficiencies. That  
 311 is, same sign (opposite sign) injection and detection efficiencies correspond to a positive  
 312 (negative)  $\Delta V_{nl}$ . Microscopically, the individual signs of these efficiencies are determined

313 by the difference in the spin-resolved interface conductances  $g_{\uparrow} - g_{\downarrow}$ , where the “up” direc-  
 314 tion is defined by the energy-integrated majority spin direction (*i.e.*, magnetization) of the  
 315 ferromagnet. Because the nonlocal voltage depends on the product of the two efficiencies,  
 316 it is not possible to correlate its sign directly with the sign of the spin accumulation. At  
 317 low temperatures, the influence of the electronic Knight field on the nuclear polarization in  
 318 oblique Hanle geometries [12, 19] can be used to determine the sign of the spin accumulation  
 319 with respect to the magnetization orientation. We have determined that at high forward  
 320 bias (spin extraction) the sign of the spin accumulation is minority in  $\text{Co}_2\text{FeSi}$  and majority  
 321 in  $\text{Co}_2\text{MnSi}$  with respect to the magnetization of the injector contact [55].

## 322 B. Injector-detector separation dependence

323 We quantify device parameters at different temperatures using the injector-detector sep-  
 324 aration dependence (IDSD) of the spin valve signal size, rather than relying on NLH mea-  
 325 surements. The NLH measurement in  $n$ -GaAs becomes challenging at high temperatures  
 326 because of the magnetoresistance backgrounds present over the much larger magnetic field  
 327 range required when the spin lifetime is small. The injector-detector separation was varied  
 328 in order to extract the spatial dependence of the spin accumulation in the channel. By uti-  
 329 lizing the enhanced signal in the BDSV configuration, clear SV signals could be measured at  
 330 the smallest separations up to room temperature. For the IDSD measurement, the detector  
 331 contact forward bias was fixed at a current density of  $40 \text{ A/cm}^2$ . This bias current was well  
 332 into the enhancement regime shown in Fig. 5(c), but below the regime where spin drift  
 333 enhancements were significant at low temperatures.  $\Delta V_{nl}$  was recorded at bias conditions  
 334  $J_i = 1000 \text{ A/cm}^2$ ,  $J_d = 40 \text{ A/cm}^2$  for each temperature and injector-detector separation.  
 335 The results of the IDSD measurement are summarized in Fig. 6. The solid lines in Fig. 6 are  
 336 fits to a numerical model of the spin accumulation in the channel, which will be explained  
 337 in detail later in this article.

339 We note that in Eq. 1,  $\Delta V_{nl}$  is proportional to the spin accumulation  $n_{\uparrow} - n_{\downarrow}$  and the  
 340 inverse compressibility of the channel  $\partial\mu/\partial n$ . At temperatures above the Fermi temperature  
 341 (in our samples  $T_F \simeq 60 \text{ K}$ ) at which the  $n$ -GaAs is no longer degenerate,  $\partial\mu/\partial n$  is a function  
 342 of temperature. In the nondegenerate regime ( $T \gg T_F$ ),  $\partial\mu/\partial n \propto T$ . This relationship  
 343 implies that as the temperature increases in the nondegenerate regime, a larger  $\Delta V_{nl}$  is



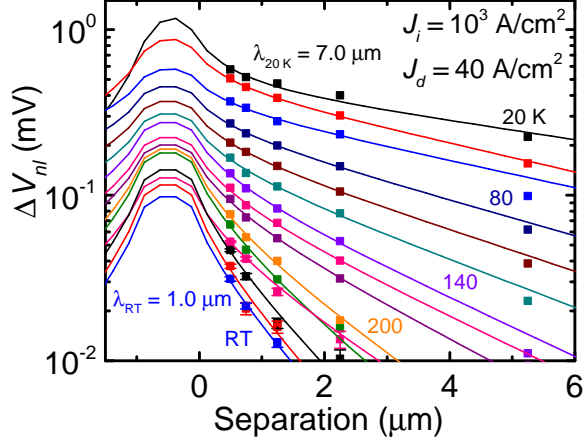


FIG. 6. (Color online) The injector-detector separation dependence of  $\Delta V_{nl}$  for the devices with  $\text{Co}_2\text{FeSi}$  contacts at temperatures from 20 K to 300 K, in increments of 20 K. The horizontal axis of the plot is the injector edge to detector center separation, *i.e.* the 1  $\mu\text{m}$ -wide injector extends from -1 to 0  $\mu\text{m}$  on the horizontal axis. Superimposed as solid lines are the fits of a 2D numerical solution of Eq. 5 with  $\tau_s$  and  $\eta\alpha$  as the fitting parameters. The bias conditions are indicated on the figure as well as the spin diffusion lengths at 20 K and room temperature (RT). At low temperature, the IDSD measurement on the  $\text{Co}_2\text{MnSi}$  devices yielded comparable SV signal sizes and  $n$ -GaAs spin diffusion length. A complete temperature-dependence measurement, however, was not performed.

344 measured *for a given spin accumulation*. For these samples,

$$\left. \frac{\partial \mu}{\partial n} \right|_{300 \text{ K}} \simeq 7 \left. \frac{\partial \mu}{\partial n} \right|_{20 \text{ K}}. \quad (3)$$

345 Because of this enhancement factor, while the spin accumulation falls by two orders of  
 346 magnitude from 20 K to 300 K,  $\Delta V_{nl}$  at separations much smaller than a diffusion length  
 347 only decreases by roughly one order of magnitude over the same temperature range.

### 348 C. Modeling of the spatial decay of spin accumulation

349 Here we discuss the model used to describe the spin accumulation in the channel and  
 350 which is used to fit the IDSD measurement results. Typically, in systems where spin diffusion  
 351 is one-dimensional, the SV signal size is interpreted with the expression [35]

$$\Delta R_{nl} = \Delta V_{nl}/I = \frac{\eta^2 \rho \lambda e^{-y/\lambda}}{A}, \quad (4)$$

352 where  $\rho$  is the channel resistivity,  $A$  is the channel cross-sectional area, and  $y$  is injector-  
 353 detector separation. Eq. 4 has been used to model the SV signal size in a variety of material  
 354 systems [1, 6, 39] in which the FM/NM barrier resistance is much larger than the channel  
 355 spin resistance, so that the conductivity mismatch problem [21] may be ignored. We choose  
 356 to use a more general numerical model of the spin accumulation in the channel to fit to  
 357 the IDSD measurement because of several considerations. First, as discussed earlier, drift  
 358 due to the bias current influences the spatial spin accumulation profile in  $n$ -GaAs at low  
 359 temperatures, and the exact drift field is best captured by a numerical model. Second, at  
 360 measurement temperatures near room temperature the spin diffusion length in  $n$ -GaAs is  
 361 less than the channel thickness of  $2.5 \mu\text{m}$ . In this regime a more general solution of the  
 362 spin drift-diffusion equation is needed, because Eq. 4 is only appropriate for devices where  
 363 the spin drift and diffusion are effectively one dimensional. In two or three dimensions, the  
 364 spin accumulation decays faster than  $e^{-y/\lambda}$  for  $y < \lambda$ , in exact analogy to the two and three  
 365 dimensional solutions of the screened Poisson equation.

366 The spatial profile of spin accumulation in the channel is modeled by solving the spin  
 367 drift-diffusion equation [40] in steady state,

$$\frac{\partial \mathbf{P}}{\partial t} = 0 = -\frac{\mathbf{P}}{\tau_s} + D\nabla^2 \mathbf{P} + \frac{\mathbf{J}}{ne} \cdot \nabla \mathbf{P} + \frac{\alpha \hat{\mathbf{m}}_i |\mathbf{J}_i|}{ne\Delta z}, \quad (5)$$

368 where  $|\mathbf{P}| \equiv (n_\uparrow - n_\downarrow)/n$  is the dimensionless spin polarization of the channel,  $D$  is the  
 369 spin diffusion constant (equal to the charge diffusion constant [40]),  $\hat{\mathbf{m}}_i$  specifies the injector  
 370 contact magnetization direction, and the last term specifies the source term, which is only  
 371 nonzero at the cells of the finite element model where spin injection occurs. In the source  
 372 term, the  $\Delta z$  factor in the denominator is the size of the injection cell in the  $z$ -direction,  
 373 which normalizes the injection rate in the finite-element grid properly.  $\mathbf{J}$  is the current  
 374 density in the channel, and the parameter  $\alpha$  is the spin injection efficiency at the FM/SC  
 375 interface (*i.e.* for  $\alpha = 1$  the spin current at the FM/SC interface is equal to the charge  
 376 current).  $\alpha$  encompasses both the bulk polarization of the current in the FM, as well as  
 377 interface effects determining the polarization of the charge current. The spin valve device  
 378 geometry is cast into a finite-element grid, and Eq. 5 is solved numerically by forward  
 379 iteration until steady state is reached. See Fig. 7 for a schematic diagram illustrating the  
 380 model geometry. The contact length in the  $x$ -direction ( $50 \mu\text{m}$ ) is much longer than the  
 381 spin diffusion length at all temperatures. The model is therefore confined to the  $yz$ -plane  
 382

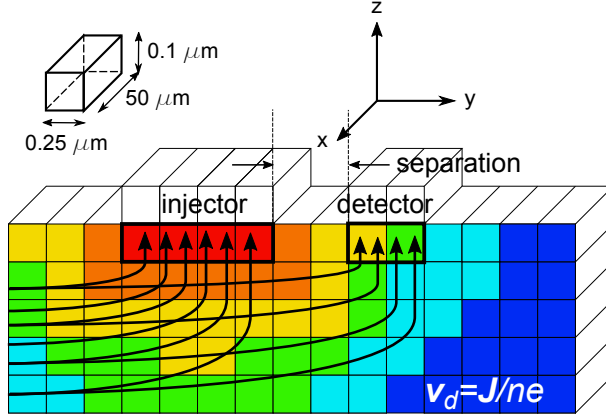


FIG. 7. (Color online) Schematic illustrating the 2D finite-element model used to solve Eq. 5 numerically. The spin accumulation, which drifts and diffuses from the injector contact, is indicated for illustrative purposes in false color (red high, blue low). The channel drift velocity  $\mathbf{v}_d = \mathbf{J}/ne$  is schematically shown by the field lines. The bolded black outlines the cells in which injection and detection occurs. The cell dimensions  $\Delta x, \Delta y, \Delta z$  used in the simulation are shown in the upper left. The number of cells drawn is not the actual number of cells used, nor is the model drawn to scale.

383 and the spin accumulation is assumed to be uniform in the  $x$ -direction. Neumann boundary  
 384 conditions are enforced at the free boundary cells, *i.e.* the diffusive spin current  $\propto \nabla \mathbf{P} = 0$   
 385 at the boundaries.

386 The current density  $\mathbf{J}$  in the channel was solved for prior to solving Eq. 5 by assuming  
 387 charge neutrality throughout the channel, so that  $\nabla \cdot \mathbf{E} = \nabla \cdot \mathbf{J} = 0$ . Because  $\nabla \cdot \mathbf{J} = 0$ ,  
 388 there exists a scalar potential  $\phi_J$  that satisfies  $\nabla^2 \phi_J = 0$ .  $\phi_J$  is solved for with a Laplace  
 389 relaxation method, and finally the current density vector field is solved for by evaluating  
 390  $\nabla \cdot \phi_J = \mathbf{J}$ .

391 The diffusion constant  $D$  is calculated from the Einstein relation

$$D = \frac{n\nu}{e} \left( \frac{\partial \mu}{\partial n} \right), \quad (6)$$

392 where  $\nu$  is the mobility. For  $n = 2.8 \times 10^{16}$  GaAs, the Fermi temperature  $T_F \simeq 60$  K, so  
 393 in order to capture the transition from degenerate to nondegenerate behavior, the inverse  
 394 compressibility  $\partial \mu / \partial n$  is calculated using full Fermi-Dirac statistics. A parabolic conduction  
 395 band density of states with GaAs effective mass  $m^* = 0.067m_0$  [28] is used, and the inverse

396 compressibility is evaluated via the expression

$$\frac{\partial\mu}{\partial n} = \frac{k_b T}{n} \frac{F_{1/2}(\zeta)}{F_{-1/2}(\zeta)}, \quad (7)$$

397 where  $\zeta \equiv \mu/k_b T$  is the reduced chemical potential and  $F_\alpha(\zeta)$  is the complete Fermi-Dirac  
 398 integral. In the limits  $T \ll T_F$  and  $T \gg T_F$  Eq. 7 reduces to  $\partial\mu/\partial n = 2E_F/3n$  and  
 399  $\partial\mu/\partial n = k_b T/n$ , respectively.

400 To compare the solution of Eq. 5 directly with the measured  $\Delta V_{nl}$ , the calculated nonlocal  
 401 spin accumulation at the detector is input to Eq. 1. The overall scale of  $\eta$ , the detection  
 402 efficiency, cannot be determined in this measurement. However, because the known injector  
 403 current density constrains the spin injection rate, the product of the injection and detection  
 404 efficiencies  $\eta\alpha$  can be determined. We will discuss the constraints on  $\eta$  in more detail below.

405 The IDSD measurement results are fit to the numerical solution of Eq. 5, with the spin  
 406 lifetime  $\tau_s$  and the dimensionless spin injection efficiency  $\alpha$  as fitting parameters. The fits  
 407 to the IDSD results are shown as solid lines in Fig. 6, and the temperature dependence of  
 408 the fitting parameters  $\tau_s$  and  $\eta\alpha$  are shown in Figs. 8(a) and (b). The product  $\eta P_0$  of the  
 409 detection efficiency and the spin polarization  $P_0$  below the injector is also shown in Fig. 8(b).

#### 410 **D. Hanle fitting**

411 At low temperatures, at which the NLH measurement could be performed, the spin  
 412 lifetime obtained from fits of the IDSD measurement could be compared to the spin lifetime  
 413 measured by Hanle precession experiments. To fit NLH field sweeps the data were fit to the  
 414 Green's function solution of Eq. 5 in one dimension, which gives

$$V_{nl}(H) \propto \mathbf{P}(y) \cdot \hat{\mathbf{m}}_d \propto \int_{-\infty}^t \frac{\exp[-(\frac{y^2}{4Dt} + \frac{t}{\tau_s})]}{\sqrt{4\pi Dt}} \cos(\gamma_e H t) dt, \quad (8)$$

415 where  $|\gamma_e|/2\pi = 0.62$  MHz/Oe is the gyromagnetic ratio in GaAs. Eq. 8 is identical to  
 416 solving Eq. 5 in one dimension with an added precession term from an external transverse  
 417 magnetic field  $H$ , and  $\mathbf{J} = 0$ . The simplification to one dimension is appropriate at low  
 418 temperatures, because the spin diffusion length  $\sqrt{D\tau_s}$  is larger than the channel depth of  
 419  $2.5 \mu\text{m}$ .

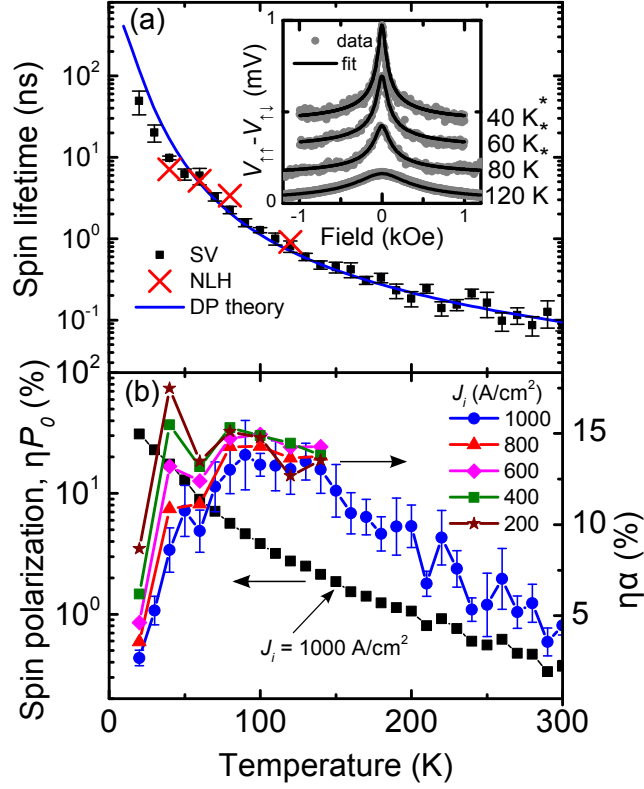


FIG. 8. (Color online) (a) The temperature dependence of  $\tau_s$  extracted from the fits in Fig. 6 along with the theoretical prediction based on Eq. 9, which is shown as the blue solid line. Spin lifetimes extracted from NLH measurements are shown as red crosses, with the corresponding NLH data  $V_{\uparrow\uparrow} - V_{\uparrow\downarrow}$  and fits to Eq. 8 shown in the inset (artificially offset). The asterisks on the temperature labels in the inset indicate that the NLH sweeps were taken with the pulsed current measurement to mitigate DNP effects. The NLH data shown are taken at the same bias currents as used for the data of Fig. 6 on the 250 nm separation device. (b) The temperature dependence of  $\eta P_0$  (left ordinate) and  $\eta\alpha$  (right ordinate).  $P_0$  is the spin polarization directly beneath the injector from the model fits shown in Fig. 6. At temperatures below 140 K,  $\eta\alpha$  is shown for different injector current densities using the symbols indicated in the legend. In (b) representative error bars are shown for the  $J_i = 10^3$  A/cm<sup>2</sup> data only. All data in (b) were taken with  $J_d = 40$  A/cm<sup>2</sup>.

#### 420 E. Spin lifetime calculation

421 In order to compare the measured temperature dependence of the spin lifetime with DP  
 422 theory, we used the method of Lau, Olesberg, and Flatté [56, 57] to calculate the spin

423 relaxation rate for the doping concentration  $n = 2.8 \times 10^{16} \text{ cm}^{-3}$ . The spin relaxation rate,  
 424  $\tau_s^{-1}$ , can be expressed as

$$\tau_s^{-1} = \frac{1}{\tilde{n}} \int D(\epsilon) f(\epsilon) [1 - f(\epsilon)] \tau_3(\epsilon) \Omega_3^2(\epsilon) d\epsilon, \quad (9)$$

425 where  $D(\epsilon)$  is the effective-mass approximation density-of-states in the GaAs,  $f(\epsilon)$  is the  
 426 Fermi-Dirac distribution function,  $\tau_3$  is the  $l = 3$  component in the multipole expansion of  
 427 the momentum scattering time, and  $\Omega_3(\epsilon)$  is the  $l = 3$  component of the energy-dependent  
 428 effective SOI magnetic field. The cubic symmetry of the Dresselhaus interaction in bulk  
 429 GaAs [15] results in  $\Omega_l^2 = 0$  for all  $l \neq 3$ . Eq. 9 is a generalization of the original DP  
 430 expression  $\tau_s^{-1} = a \langle \Omega^2 \rangle \tau_p$  [17, 19], where the integral over energy in Eq. 9 properly weights  
 431 the spin relaxation rate to account for an arbitrary degree of degeneracy as well as energy-  
 432 dependent momentum scattering mechanisms.

433 In  $n$ -GaAs, the dominant scattering mechanism changes from ionized-impurity (II) scat-  
 434 tering at low temperatures to optical-phonon (OP) scattering at high temperatures [58], as  
 435 demonstrated by the non-monotonic temperature-dependence of the mobility shown in Fig.  
 436 3(a). To determine the momentum scattering time, the experimental mobility  $\nu$  is fit to the  
 437 form

$$\nu^{-1} = \underbrace{(A + BT^{3/2})^{-1}}_{\nu_{\text{II}}} + \underbrace{(CT^{-1})^{-1}}_{\nu_{\text{OP}}}, \quad (10)$$

438 which combines the II and OP scattering rates via Matthiessen's rule. In Eq. 10, A and B are  
 439 fitting parameters for the II mechanism and C is a fitting parameter for the OP mechanism.  
 440 For II scattering,  $T^{3/2}$  is the known temperature dependence of the scattering time [59] and  
 441 the fitting parameter  $A$  is added to account for degeneracy at low temperatures. No universal  
 442 energy exponent can be assigned to OP scattering over the experimental temperature range,  
 443 due to the breakdown of the relaxation-time approximation [58, 60]. We find, however, that  
 444  $\nu \propto T^{-1}$  approximates the measured high temperature mobility. This is not a rigorous  
 445 relation for OP scattering, but the purpose of Eq. 10 is to provide a phenomenological  
 446 scattering rate which *decreases* with temperature (II scattering) and a scattering rate which  
 447 *increases* with temperature (OP scattering). The fit to Eq. 10 is shown along with the  
 448 measured mobility in Fig. 3(a).

449 After fitting the temperature dependence of the mobility to extract the contributions due

450 to the II and OP scattering mechanisms, each mechanism is separately fit to the expression

$$\nu_{\text{II(OP)}} = \frac{e}{m^*n} \int D(\epsilon) f(\epsilon) [1 - f(\epsilon)] \tau_{1,\text{II(OP)}}(\epsilon) \frac{\epsilon}{k_b T} d\epsilon \quad (11)$$

451 to determine  $\tau_1$  (the momentum relaxation time) for each mechanism, at each temperature.  
 452 The energy dependence of the scattering time is assumed to be  $\tau_1 = a\epsilon^\gamma$ , where  $\gamma = 3/2$   
 453 and  $\gamma = 1/2$  for II and OP scattering, respectively [57]. The relevant multipole component  
 454 of the scattering time for DP relaxation,  $\tau_3$ , can be determined from  $\tau_1$  by expressing the  
 455  $l^{\text{th}}$  multipole component of the scattering time using the known form of the scattering cross  
 456 section  $\sigma(\theta, \epsilon)$

$$\tau_l^{-1}(\epsilon) = \int_0^\pi \sigma(\theta, \epsilon) [1 - P_l(\cos\theta)] \sin\theta d\theta, \quad (12)$$

457 where  $P_l$  is the Legendre polynomial of degree  $l$ . Eq. 12 may be evaluated to relate  $\tau_3$  to  $\tau_1$   
 458 (for detailed evaluation of Eq. 12 see Ref. 19, resulting in  $\tau_1 = \tau_3/6$  for II scattering, and  
 459  $\tau_1 = 6\tau_3/41$  for OP scattering [19, 57]).

460 After fitting the measured mobility with Eq. 10 and 11, the  $l = 3$  component of the  
 461 momentum scattering rate  $\tau_3^{-1} = \tau_{3,\text{II}}^{-1} + \tau_{3,\text{OP}}^{-1}$  is input to Eq. 9, and the DP spin relaxation  
 462 rate is evaluated at all temperatures. The SOI strength used to evaluate  $\Omega_3^2$  as a function  
 463 of carrier energy is taken from the  $k \cdot p$  calculation with a full fourteen band basis done by  
 464 Lau *et al.* [56]. Their calculations give  $\mathbf{\Omega} = 2\beta/\hbar(\mathbf{k}_x(k_y^2 - k_z^2) + \mathbf{k}_y(k_z^2 - k_x^2) + \mathbf{k}_z(k_x^2 - k_y^2))$   
 465 with  $\beta = 25 \text{ eV \AA}^3$ . The final result for the spin lifetime as a function of temperature from  
 466 Eq. 9 is shown as the blue solid line in Fig. 8(a).

#### 467 IV. DISCUSSION

468 As shown in Fig. 6, the spin diffusion length  $\lambda = \sqrt{D\tau_s}$  falls from approximately  $7 \mu\text{m}$  at  
 469 20 K to  $1 \mu\text{m}$  at room temperature. Injector-detector separations less than approximately  
 470  $1.0 \mu\text{m}$  are therefore ideal to detect NLSV signals in  $n$ -GaAs at room temperature. We  
 471 emphasize that a two-dimensional model of spin diffusion is needed to fit the separation  
 472 dependence of  $\Delta V_{nl}$  when the spin diffusion length is smaller than the channel depth of  $2.5$   
 473  $\mu\text{m}$ . Fits using the 1D solution of Eq. 5 underestimate the spin lifetime and spin diffusion  
 474 length when the channel thickness is greater than a spin diffusion length, because the spin  
 475 accumulation in two dimensions decays faster than  $e^{-y/\lambda}$  away from the injector.

476 As can be seen in Fig. 8(a), the temperature dependence of the spin lifetime agrees well  
 477 with the DP prediction, calculated from Eq. 9, over the entire temperature range.  $\tau_s$  varies  
 478 from  $49 \pm 16$  ns at 20 K to  $86 \pm 10$  ps at 300 K. The relatively large uncertainty in the 20 K  
 479 spin lifetime value results from a lack of data for injector-detector separations larger than the  
 480 spin diffusion length at low temperature. Separations larger than  $10 \mu\text{m}$  would be required  
 481 to constrain the fit adequately. At low temperatures (40-120 K) we have also measured  $\tau_s$   
 482 by the NLH measurement. The spin lifetimes obtained with NLH measurements are also  
 483 shown on Fig. 8(a), with the NLH field sweeps and fits to Eq. 8 shown in the inset. The  $\tau_s$   
 484 values from NLH measurements are in good agreement with the IDSD  $\tau_s$  values above  $\sim 60$   
 485 K. At the lowest temperatures (20-40 K), the pulsed NLH measurement technique may not  
 486 be sufficient to completely remove the effects of DNP. A combined model of the electron-  
 487 nuclear spin system is needed to adequately model the NLH measurement in the regime  
 488 where DNP is significant, as is done in Refs. [12, 14, 61].

489 We now comment on the magnitude of  $\Delta V_{nl}$  in the biased-detector SV measurement.  
 490 Combining Eq. 1 and Eq. 7 allows one to determine the spin accumulation  $n_\uparrow - n_\downarrow$  given  
 491  $\Delta V_{nl}$ , the SV signal size. The only unknown is  $\eta$ , the detection efficiency. In our devices,  
 492 we have demonstrated that  $\eta$  is a strong function of detector bias, which complicates the  
 493 interpretation. Because of the detector bias dependence of  $\eta$  implied by the data shown in  
 494 Fig. 5, we also cannot assume  $\alpha = \eta$ , as the injector contact is biased with a large current,  
 495 while the detector bias is varied. Based on these considerations, the spin polarization of the  
 496 channel and the injection efficiency may only be quantitatively evaluated up to a factor of  
 497  $\eta$  (*i.e.*  $\eta P_0$  and  $\eta\alpha$ , respectively), where  $\eta$  is the detection efficiency at the detector bias  
 498 voltage at which the measurement was performed and  $P_0$  is the spin polarization below the  
 499 injector. These quantities are shown in Fig. 8(b). Although the overall scale for  $\eta$  cannot be  
 500 determined in this experiment, it is believed to be  $\sim 50\%$  based on spin-LED measurements  
 501 on similar Fe/GaAs Schottky interfaces [62].

502 At the lowest temperatures, we measure  $\Delta V_{nl}$  values of  $\sim 1$  mV with a forward bias  
 503 applied to a detector contact. This implies that the spin-resolved electrochemical potential  
 504 splitting at the injector is comparable to the Fermi energy in the GaAs channel, which is  
 505  $\sim 5$  meV with respect to the conduction band minimum. As the maximum possible value of  
 506  $\eta$  is unity, we emphasize that the ordinate scales shown in Fig. 8(b) are therefore minimum  
 507 values for  $P_0$  and  $\alpha$ . At 20 K, we measure  $\eta P_0 = 30\%$ . Thus, the upper limit of 100%



508 polarization in the GaAs puts a *lower* limit of  $\eta \sim 0.3$  at 20 K. Notably, because the  
 509 forward bias current (spin extraction) leads to drift *enhancement* of the spin accumulation  
 510 buildup at the injector contact, ideal ferromagnetic contacts ( $\alpha = 1$ ) are not necessary to  
 511 achieve channel spin polarizations approaching 100% [40, 63].

512 In Fig. 8(b), a downturn in the injection-detection efficiency product  $\eta\alpha$  is observed at  
 513 temperatures below 100 K. To address this observation, we have measured  $\eta\alpha$  for different  
 514 injector current biases. The results of this measurement are shown in Fig. 8(b), where  
 515 it is apparent that  $\eta\alpha$  is a function of the injector current bias at low temperatures. At  
 516 temperatures above  $\sim 150$  K, where the spin accumulation is small with respect to the  
 517 carrier density,  $\eta\alpha$  becomes independent of injector current bias.

518 To understand the injector bias current dependence of  $\eta\alpha$ , we first discuss the influence  
 519 of an electric field on the spin accumulation. Electric fields at the injector necessarily ac-  
 520 company the bias current. In addition to the drift effects, discussed above, large electric  
 521 fields in *n*-GaAs are known to enhance the spin relaxation rate. In *n*-GaAs, at low tem-  
 522 peratures ( $T \lesssim 30$  K) the itinerant electron temperature can deviate significantly from the  
 523 lattice temperature due to the dominance of elastic scattering mechanisms, which hinder  
 524 electron-lattice equilibration [64]. This electron heating is present above electric fields  $\sim 10$   
 525 V/cm, and leads to donor impact ionization, which prevents the electron temperature from  
 526 cooling below the donor binding energy ( $\sim 6$  meV for Si in GaAs [28]). At low temperatures,  
 527 electric field dependence of the spin lifetime has been widely reported [9, 65, 66]. At the  
 528 lowest temperatures in our experiment (20, 30 K), the suppression of the spin lifetime due  
 529 to the applied electric field may contribute to the downturn in  $\eta\alpha$  we observe. However,  
 530 the injector bias dependence of  $\eta\alpha$  is observed clearly up to  $\sim 100$  K in Fig. 8(b). At 100  
 531 K, all donors are thermally ionized and inelastic electron-phonon relaxation mechanisms  
 532 are sufficient to prevent any electron-lattice temperature difference. Thus, we believe that  
 533 electric field suppression of the spin lifetime is not the origin of the injector bias dependence  
 534 of  $\eta\alpha$ .

535 We believe that the downturn in  $\eta\alpha$  at low temperatures is more likely to be a consequence  
 536 of the large spin polarization of the channel and consequent breakdown of the ordinary  
 537 drift-diffusion model. In the presence of a spin accumulation comparable to the carrier  
 538 density, Eq. 5 must be modified to prevent the spin polarization from achieving non-physical  
 539 values  $> 100\%$ . Physically, the model parameters themselves become functions of the spin

540 polarization, and the assumption of linear response breaks down [67]. To be specific, it  
 541 becomes necessary to specify the diffusion constants and spin relaxation rates separately for  
 542 minority and majority spin carriers, *i.e.*  $\tau_{\uparrow\downarrow}^{-1} \neq \tau_{\downarrow\uparrow}^{-1} \neq \tau_{s,0}^{-1}/2$  and  $D_{\uparrow} \neq D_{\downarrow} \neq D_0$ , where  $\tau_{s,0}^{-1}$   
 543 and  $D_0$  are the equilibrium spin relaxation rate and diffusion constant, respectively [68]. We  
 544 note that for the DP spin relaxation mechanism ( $\tau_s^{-1} \sim \epsilon^3 \tau_p$ ) in *n*-GaAs where II scattering  
 545 is dominant ( $\tau_p \sim \epsilon^{3/2}$ ) the spin relaxation rate is a strong function of carrier energy  $\epsilon$ .  
 546 The diffusion constant also increases with increasing carrier energy via the Einstein relation  
 547 (Eq. 6). The mechanisms described above may provide feedback to limit the spin polarization  
 548 in the large spin polarization regime via more efficient spin diffusion and spin relaxation  
 549 processes compared to the small spin polarization linear-response limit. If this were the  
 550 case, then the injector current polarization required to achieve a given spin accumulation  
 551 would be larger than that calculated under the assumption of linear response.

## 552 V. CONCLUSIONS

553 In conclusion, we have explored several aspects of spin transport in epitaxial FM/*n*-GaAs  
 554 spin valves over a wide range of temperature and bias conditions. Because these devices are  
 555 based on Schottky tunnel barriers, both the injection and detection efficiencies depend on  
 556 the bias. We have exploited this property to enhance the sensitivity to spin accumulation by  
 557 applying a bias current to the detector in the nonlocal configuration. Although the mecha-  
 558 nism for the enhancement is not well-understood (except for the role of drift), this approach  
 559 enables detection of spin accumulation up to room temperature. At injector current densi-  
 560 ties of  $10^3$  A/cm<sup>2</sup> nonlocal voltages of order  $\sim 1$  mV are detected at low temperature, which  
 561 fall to  $\sim 40$   $\mu$ V at room temperature. This approach has enabled measurements of the spin  
 562 relaxation rate and diffusion length over the entire temperature range, and good agreement  
 563 is obtained with a model based on the Dyakonov-Perel spin relaxation mechanism. At the  
 564 lowest temperatures, however, the standard drift-diffusion model appears to break down  
 565 because of the large spin accumulation, which is comparable to the carrier density. At high  
 566 temperatures, the devices are limited by the rapidly increasing spin relaxation rate, although  
 567 the injected current polarization also decreases by a factor of three between 20 K and room  
 568 temperature.

569 The devices discussed in this paper are based on Heusler alloys, which are predicted to

570 have a high spin polarization and grow epitaxially on GaAs (001). There is sufficient un-  
571 certainty in the derived values of the detection efficiency and injected current polarization  
572 that it is not possible to make a statement about the polarization of the Co<sub>2</sub>FeSi injector  
573 beyond the lower bound (30%) set by the size of the nonlocal voltage at the lowest tem-  
574 perature. As suggested by the bias dependence, there is likely a significant contribution  
575 to the tunnelling current from interface states, a property that is shared by the epitaxial  
576 Fe/GaAs system [54]. Although these important details still need to be resolved, this work  
577 demonstrates that epitaxial FM/III-V heterostructures can be used to probe spin transport  
578 at room temperature.

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