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Spin relaxation in a germanium nanowire

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We report experimental study of spin transport in nanowire spin valve structures consisting of three layers—cobalt, germanium, and nickel. The spin diffusion length in the Ge is estimated to be about 400 nm at 1.9 K and the corresponding spin relaxation time is about 4 ns. At 100 K, the spin diffusion length drops to 180 nm and the relaxation time is about 0.81 ns. These short relaxation times, which depend weakly on temperature, are caused by strong surface roughness scattering that causes rapid spin relaxation via the Elliott-Yafet mode [Elliott, Phys. Rev. **96**, 266 (1954)]. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2230012]

I. INTRODUCTION

Germanium is an elemental semiconductor that possesses crystallographic inversion symmetry. As a result, the Dresselhaus spin orbit interaction,[1] a major cause of spin relaxation in solids, is nominally absent. Normally, this should lead to long electron spin relaxation times in a germanium sample, but if there is any external electric field breaking structural inversion symmetry, then the resulting Rashba spin orbit interaction[2] will cause some spin relaxation via the D'yakonov-Perel'[3] (DP) and Elliott-Yafet[4] (EY) modes.

Both the DP and the EY relaxation mechanisms have a strong dependence on dimensionality. However, these dependences are *opposite* for the two mechanism. The DP mechanism is suppressed by quasi-one-dimensional confinement,[5–7] while the EY mechanism may be enhanced because of an increase in the momentum relaxation rate due to frequent surface roughness scattering. Thus, we expect the EY mechanism to predominate in a quasi-one-dimensional system such as a nanowire. Furthermore, if the momentum relaxing scattering events causing the EY spin relaxation are mostly elastic (such as surface roughness scattering), then we expect the momentum relaxation rate to be relatively temperature independent. In that case, the spin relaxation rate will be relatively temperature independent as well, since in the EY mechanism, the spin relaxation rate has the same temperature dependence as the momentum relaxation rate.[8–10]

In this paper, we report experimental study of spin relaxation in Ge nanowires of 50 nm diameter. The nanowires are embedded in an insulating matrix that has a strong built-in electric field due to fixed charges resident in the insulator.[11] This electric field gives rise to Rashba spin orbit interaction, which causes spin relaxation.

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II. FABRICATION OF NANOWIRE SPIN VALVES

The popular structure used to study spin relaxation in any paramagnetic material is the "spin valve." It consists of a paramagnetic layer flanked by two ferromagnetic contacts acting as spin injector and spin detector. We have fabricated nanowire spin valve structures, each consisting of a Ge spacer between Co and Ni contacts. The nanowires have a diameter of 50 nm. The devices are self-assembled using electrochemistry. We start with a high purity (99.997%) metallic aluminum foil (0.1 mm thick), which is electropolished in a suitable organic solution[12] to produce a mirror-like surface. An anodic alumina film with highly ordered nanopores is then formed on this electropolished surface by a multistep anodization procedure.[13] The anodization conditions (e.g., the nature of the acidic electrolyte, anodization voltage, etc.) determine the dimensions of the nanopores. In this work we have used 0.3M oxalic acid as the electrolyte, and the anodization voltage has been kept constant at 40 V dc. Under these conditions we get a porous alumina film forming on the surface of the aluminum which has a nominal pore diameter of 50 nm. Figure 1(a) shows a scanning electron micrograph of the pores viewed from the top. Anodization was carried out for 10 min which results in an alumina film of thickness of ~ 1 μm, so that the pores are ~ 1 μm deep. There is a 40–50 nm thick alumina "barrier layer" at the bottom of the pores [see Fig. 1(b)] which is a barrier to current flow along the length of the pores. This layer is removed by a "reverse polarity etching" technique[14] using a dc voltage of 35 V. Figure 1(c) shows the etched porous film viewed from the bottom after stripping off the Al foil in $HgCl_2$ solution. Note that reverse polarity etching has opened up some (but not all) pores from the bottom, meaning that the barrier layer has been successfully removed in some cases.

We synthesize the spin valves by sequentially electrodepositing Ni, Ge, and Co inside the pores. Electrodepositions of Ni and Co are carried out in dilute aqueous solutions of the metal-sulfate salt with slightly acidic pH. A small dc bias of +5 V is applied at a platinum counter electrode with respect to the aluminum foil. Since the Al foil is at a negative

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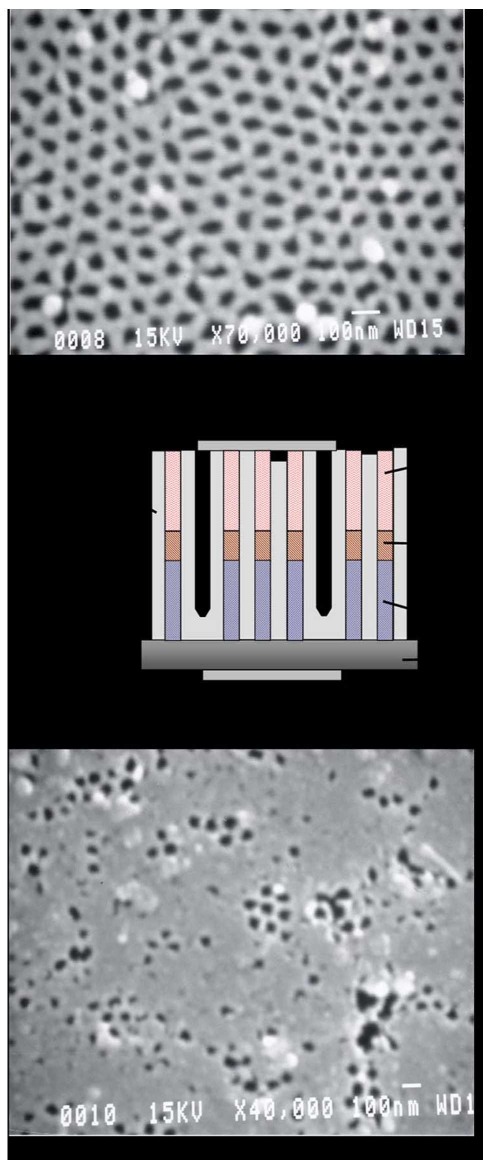

FIG. 1. (Color online) (a) Scanning electron micrograph of the pores (in the alumina film) viewed from the top; (b) side view of the structure showing the trilayered nanowire spin valves hosted in the pores. Note that only few wires will be electrically contacted from both ends and will be probed; (c) scanning electron micrograph of the porous film viewed from the bottom after the reverse polarity etching procedure and after the Al foil has been dissolved in $HgCl_2$ solution. Note that some, but not all, pores have been opened up from the bottom.

potential, the Co^{++} or Ni^{++} ions in solution preferentially electrodeposit within the pores, since they offer the least resistance path for the current to flow.

Small deposition current (approximately microamperes) ensures slow and well-controlled electrodeposition of metals inside the pores. We calibrated the deposition rate of each metal. To achieve this, we monitored the deposition current during electrodeposition of each metal inside an anodic alumina film of known pore length. The deposition current increases drastically when the pores are completely filled up. The average deposition rate is then determined by calculating the ratio of pore length to pore filling time. According to this calibration, thicknesses of Ni and Co layers in the spin valves are estimated to be ~300 and ~500 nm, respectively.

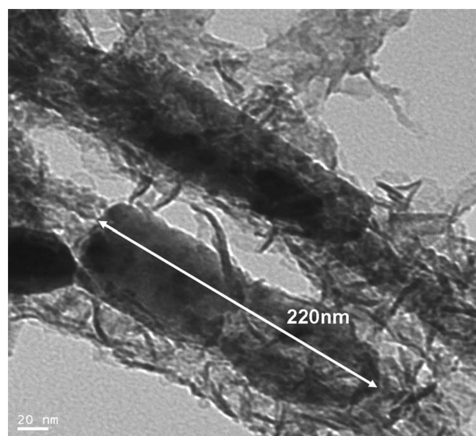

FIG. 2. Transmission electron micrograph of two released nanowires showing that the thickness of the Ge spacer is about 220 nm. This length agrees very well with what we estimated (200 nm). The Ge layer thickness is also quite uniform, since both wires shown in this micrograph have nearly the same Ge layer thickness.

Germanium is electrodeposited within the pores following the method of Refs. 15 and 16. We use an electrolyte consisting of a mixture of an ionic liquid 1-butyl-3-methyl-imidazolium-hexafluorophosphate ($BMIPF_6$), GeI_4 crystals, and 250 ml of dimethyl sulfoxide (DMSO) at concentrations of about $10^{-2}M$. GeI_4 does not dissolve in water but dissolves in this solution. In order to increase the conductivity of the electrolyte, a small amount of lithium perchlorate is added. The mixture is stirred overnight and electrodeposition is carried out at 70 °C with a 3 V dc bias for 6–8 s. The samples are then rinsed in de-ionized (DI) water and annealed at about 200 °C for 12 h to establish a good Ohmic contact between the metal and Ge surfaces.

Since Ge electrodeposition is not well calibrated, we could not estimate the thickness of the Ge layer from the duration of electrodeposition. Therefore, we dissolved the alumina host matrix in NaOH solution, centrifuged the solution to collect the released wires at the bottom of the solution, dispersed the wires in alcohol by sonification, and then captured them on transmission electron microscopy (TEM) grids for imaging. Figure 2 shows a TEM micrograph of two captured wires. The Ge layer is found to have a thickness of 220 nm. Since the layer thickness is approximately the same in both randomly captured wires, we believe that the Ge layer has a relatively uniform thickness. Note that the total length of a nanowire is 300 nm(Ni)+220 nm(Ge) +500 nm(Co)=1.02 μm, which is very close to the estimated pore length of ~1 μm.

After the completion of the electrodeposition steps, we are left with a two-dimensional array of trilayered nanowires vertically standing in an insulating alumina matrix. This is shown in Fig. 1(b). The surface of the sample is then slightly etched in 0.2M chromic and 0.4M phosphoric acid at 70 °C in order to expose the tips of the *tallest* nanowires for electrical contact. A very small fraction of the nanowires is exposed by this process. At the bottom, again only a fraction of the nanowires will make contact with the Al, since reverse polarity etching does not remove the entire barrier layer *uniformly* as can be clearly seen in Fig. 1(c). Therefore, only a

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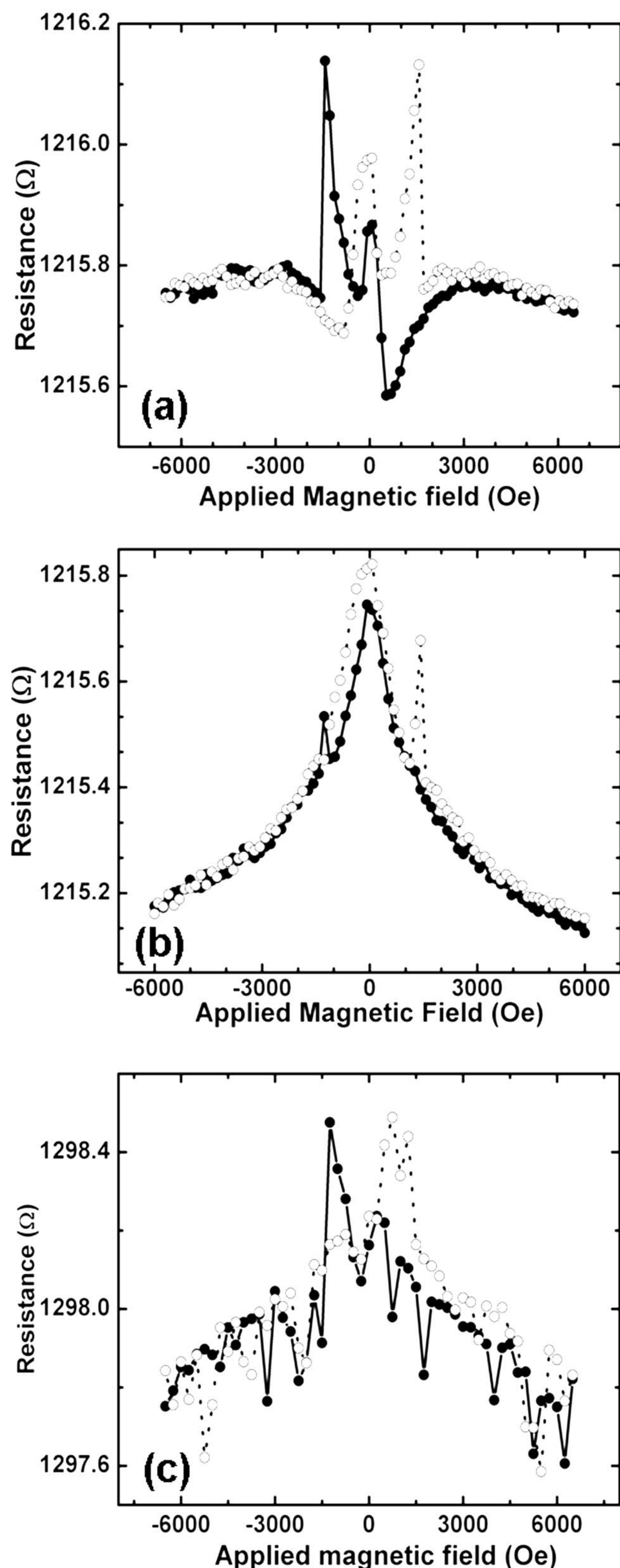

FIG. 3. Magnetoresistance of the spin valves measured at (a) 1.9 K, (b) 5 K, and (c) 100 K. The "spin valve peaks" are visible against a background negative magnetoresistance caused by the anisotropic magnetoresistance effect. The solid lines correspond to downscan when the magnetic field is swept from positive to negative values, and the broken lines correspond to upscan when the magnetic field is swept in the reverse direction. The spin valve peaks are identified with arrows.

miniscule fraction of the nanowires will be electrically contacted from *both* top and bottom. In the past, we showed that the probability of being contacted from both ends is ~ 1 in 10^8.[17] Thus, on the average, only about 1 in 10^8 nanowires is electrically probed. This is fortuitous since it allows us to measure a few nanowires using relatively large area contact pads. Finally, contacts are made to the top (Co layer) and bottom (Al) using silver paste and gold wires. The contact areas are $\sim 1 \times 1$ mm.

III. RESULTS AND DISCUSSION

Magnetoresistance of the nanowire spin valves is measured in a Quantum Design physical property measurement system. Sample temperature is varied in the range of 1.9–100 K while a magnetic field is applied along the length of the nanowires. In the experiments, the field is swept from −6 to +6 kOe. Magnetoresistance is measured with a constant current source of 10 μA.

We first note that the sample resistance is fairly temperature independent and varies from 1215 Ω at 1.9 K to 1298 Ω at 100 K. This immediately tells us that the Ge layer is *not* behaving as an intrinsic semiconductor whose carrier concentration should vary with temperature as $\sim \exp[-E_g/2kT]$ (E_g=band gap). Presumably, the fabrication process creates numerous surface states at the interface of the Ge nanowires and alumina. These states pin the Fermi level well above the conduction band, making the electron concentration in Ge very large (metal-like) and also temperature independent. Nanowires have a large surface to volume ratio, and therefore the surface states have a major effect on carrier concentration. These surface states are electrically charged and therefore result in much increased Coulomb scattering (a strong momentum relaxing mechanism), which becomes a major contributor to the EY spin relaxation. The resistance of the device actually increases slightly with increasing temperature, rather than decreasing. This is metallic behavior, and we believe that it is due to phonon scattering increasing with increasing temperature.

The measured magnetoresistance data at temperatures of 1.9, 5, and 100 K are shown in Figs. 3(a)–3(c). There is a background negative magnetoresistance which is caused by the anisotropic magnetoresistance (AMR) effect frequently observed in these structures.[18] Reference 19 used a nonlocal measurement geometry to eliminate the AMR effect, but our samples are not amenable to such procedures. In spite of the large AMR effect (1% in the field range of 0–0.6 T), we can clearly see tell-tale resistance peaks whose leading and trailing edges occur at fields approximately corresponding to the coercive fields of the Co and Ni nanomagnets.[20,21] These peaks are undoubtedly caused by the "spin valve effect." Between the coercive fields of Co and Ni, the magnetizations of the two ferromagnets are antiparallel. Therefore, one contact injects spins of a particular polarization and the other contact blocks them. Consequently, the device resistance is high. Outside this field range, the magnetizations are parallel so that the device resistance is low. This causes the resistance peak.

The magnetoresistance data become noisy at 100 K

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probably because of thermally activated traps causing telegraph noise. Nonetheless, vestiges of the spin valve peak are still discernible at this temperature. We point out that even though the trailing edges of the peaks appear to be discontinuous, these peaks are not "jumps" associated with magnetization switching that are sometimes observed in ferromagnetic nanowires.[22] The jumps are "irreversible," meaning that the magnetoresistance before the jump is never recovered after the jump. That is not the case with the peaks; the magnetoresistance before the leading edge is recovered after the magnetic field sweeps past the trailing edge of the peak. In other words, the effect is "reversible," as expected of the spin valve phenomenon.

It is possible to estimate the spin relaxation length in the Ge spacer from the measured resistance change ΔR associated with the spin valve peak. We follow the model of Refs. 18 and 19 modified for the classical spin valve geometry:

$$\Delta R = \frac{2\alpha_F^2(\lambda_N/\sigma_N A)e^{(-L/2\lambda_N)}}{(M+1)[M\sinh(L/2\lambda_N)+\cosh(L/2\lambda_N)]}, \tag{1}$$

where $M=\lambda_N\sigma_F(1-\alpha_F^2)/\lambda_F\sigma_N$, $\alpha_F=(\sigma_\uparrow-\sigma_\downarrow)/(\sigma_\uparrow+\sigma_\downarrow)$ $\equiv$ bulk current spin polarization of the ferromagnetic electrodes (assuming they are made of same material), $\sigma_\uparrow$ ($\sigma_\downarrow$) indicates the spin up (down) conductivity of the ferromagnet, σ_N (σ_F) denotes the total conductivity of the Ge (ferromagnetic) layer, λ_N (λ_F) is the spin diffusion length in the Ge (ferromagnetic) layer, $L=200$ nm is the distance between the two ferromagnetic electrodes (thickness of the Ge layer), and A is the cross sectional area through which current flows. We proceed as follows to calculate λ_N.

Consider a Ni–Ge–Co trilayered nanowire of overall length of 1.02 μm. Length of Co=500 nm, length of Ge =220 nm, and length of Ni=300 nm. We assume $\sigma_{\text{Ni}}=\sigma_{\text{Co}}=10^5\ (\Omega\ \text{m})^{-1}$. This is an order of magnitude smaller than the bulk conductivities but is appropriate for nanowires where increased surface roughness scattering degrades mobility considerably. The conductivity of Ge (σ_{Ge}) in these structures is unknown.

We will also assume $\alpha_F=0.375$ ["average" spin polarization of Ni (33%) and Co electrodes (42%)] (Ref. 23) and λ_F (spin relaxation length in the ferromagnets) =5 nm.[24]

The diameter of the trilayered nanowire is 50 nm. Thus the resistance of a single nanowire in terms of σ_{Ge} is given by

$$R_{\text{single}} = 4074.3661 + \frac{101.8592\times10^6}{\sigma_{\text{Ge}}}$$

in ohms with σ_{Ge} expressed in $(\Omega\ \text{m})^{-1}$. The first term is the resistance due to the metallic ferromagnets and the last term is the resistance of Ge.

Let us assume m nanowires are electrically contacted from both sides. The measured device resistance is $\sim1215\ \Omega$.

Thus $R_{\text{single}}/m=1215$,

TABLE I. Spin diffusion length and time in 50 nm diameter germanium nanowires at three different temperatures.

Temperature (K)	$\Delta R/R_N$ (measured)	Spin diffusion length (λ_N) (nm)	Spin diffusion time (τ) (ns)
1.9	0.000 329	400	4
5	0.000 254 2	200	1
100	0.000 230 3	180	0.81

$$\frac{4074.3661}{m} + \frac{101.8592\times10^6}{m\sigma_{\text{Ge}}} = 1215.$$

Obviously, $m>3$ because σ_{Ge} cannot be negative. If $m=4$, then $\sigma_{\text{Ge}}\sim1\times10^5\ (\Omega\ \text{m})^{-1}$. If $m=5$, then $\sigma_{\text{Ge}}=0.5\times10^5\ (\Omega\ \text{m})^{-1}$. If $m=10$, then $\sigma_{\text{Ge}}=0.12\,613\times10^5\ (\Omega\ \text{m})^{-1}$. If $m=20$, then $\sigma_{\text{Ge}}=0.050\,36\times10^5\ (\Omega\ \text{m})^{-1}$.

Case 1. $m=5$ (and higher) and $\sigma_{\text{Ge}}=0.5\times10^5\ (\Omega\ \text{m})^{-1}$ (and lower). In this case, Eq. (1) tells us that $\Delta R/R$ must saturate to a value of 1.2×10^{-4} (or less) as $\lambda_{\text{Ge}}\to\infty$. Since our measured value of $\Delta R/R$ is larger than this quantity, the only admissible values of m are $m<5$.

Case 2. Since $3<m<5$, the only admissible value is $m=4$ if $\sigma_{\text{Ge}}=1\times10^5\ (\Omega\ \text{m})^{-1}$. This means that only four nanowires were electrically connected in the experiment. The pore density is roughly $10^{10}/\text{cm}^2$ and the contact area is 1 mm^2. Therefore, 10^8 wires are covered by the contact, but only 4 of them make actual good electrical contact. This is similar to the conclusion we arrived at in Ref. 17.

Defining the resistance of the Ge layer as $R_N=L_N/\sigma_N A$, we get from Eq. (1)

$$\frac{\Delta R}{R_N} = \frac{2\alpha_F^2(\lambda_N/L_N)e^{(-L/2\lambda_N)}}{(M+1)[M\sinh(L/2\lambda_N)+\cosh(L/2\lambda_N)]} \tag{2}$$

and

$$\frac{\Delta R}{R_N} = \frac{\Delta R}{R}\frac{R}{R_N} = 5\frac{\Delta R}{R}. \tag{3}$$

Using Eqs. (2) and (3), we can find the spin diffusion length λ_N.

In calculating the spin diffusion time, we have used the relation $\lambda_N=(D\tau)^{1/2}$, where D is the diffusion constant $=(1/3)v_F^2\tau_m$. Here τ_m is the momentum relaxation time and v_F is the Fermi velocity in Ge. To calculate v_F, we use the relations $n=k_F^3/3\pi^2$, $v_F=(\hbar/2m^*)k_F$, and $\sigma_{\text{Ge}}=ne^2\tau_m/m^*$, where n is the carrier concentration in Ge, k_F is the Fermi wave vector, and m^* is the average of the longitudinal and transverse electron effective masses in Ge (=0.861 times the free electron mass).

Assuming a reasonable value for $\tau_m=10^{-14}$ s, we obtain $n=3\times10^{26}\ \text{m}^{-3}$ and $v_F=1.12\times10^5$ m/s. This means $D=4\times10^{-5}\ \text{m}^2/\text{s}$. Using this value, we have obtained the spin relaxation time τ.

Table I lists the spin diffusion lengths and spin diffusion times at three different temperatures.

Using a perturbative approach, Elliott showed that a relation can be obtained between τ_m and τ (Ref. 10) in the case of the Elliott-Yafet mechanism:

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$$\frac{\tau_m}{\tau} \propto \left(\frac{\kappa}{\Delta E}\right)^2,$$

where κ is the spin orbit coupling constant for a specific energy band (conduction band) and ΔE is the energy separation to the nearest band (valence band). Yafet has shown that the above relation is temperature independent,[9] so that the temperature dependence of τ must track the temperature dependence of τ_m since both κ and ΔE are relatively temperature independent.

We note that τ is relatively temperature independent in the range of 5–100 K, since it varies by a factor of 5 while the temperature varies by a factor of 50. Therefore τ_m must also be relatively temperature independent. Reference 8 found a T^{-5} temperature dependence of the spin relaxation time (and therefore also the momentum relaxation time) in accordance with the Bloch-Grüneisen law. Their major momentum relaxing mechanism was phonon scattering. In our case, phonon scattering is obviously not dominant since the temperature dependence is very weak. That leaves electron-electron scattering, impurity scattering, and surface roughness scattering (which includes scattering due to surface states causing a "roughness" in the surface potential) as possible mechanisms driving spin relaxation. Electron-electron scattering rate is temperature dependent and therefore can be eliminated as the major source. Consequently, we believe that impurity and surface roughness scatterings, which are both elastic mechanisms and relatively temperature independent, are the main culprits. The latter is probably the dominant mechanism, since nanowires have a very large surface to volume ratio and therefore surface roughness scattering is frequent.

IV. CONCLUSION

In conclusion, we have studied spin transport in Ge nanowires. The primary spin relaxation mechanism appears to be the Elliott-Yafet mode associated with frequent surface roughness scattering. The spin diffusion length in Ge nanowires is slightly larger than that in carbon nanotubes [~130 nm at 5 K (Ref. 25)]. Thus, Ge nanowires offer an interesting alternative to carbon nanotubes in applications involving spintronics.

ACKNOWLEDGMENTS

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